

color predominates at room temperature as well as at higher temperatures.

Prolonged heating of the yellow acetic acid (or ethanol) solution gives rise to the deep blue color observed and studied by Dimroth and co-workers.<sup>2,3</sup>

In the crystalline form both modifications decompose on standing in air (the yellow form apparently more readily) to give a brown oil having the odor of pyridine. From one such sample was isolated a small quantity of 4,4'-dipyridyl as its crystalline hydrate, m. p. 105-106°; m. p. of picrate 252-254° (Dimroth and Heene<sup>2</sup> have reported the isolation of 4,4'-dipyridyl from air-oxidized ethanolic solutions).

**Magnetic Susceptibility.**—The measurements were kindly carried out by Mr. Clayton Callis using the apparatus described by Driggs and Hopkins.<sup>12</sup> All measurements were taken at 25° on the two solid forms of N,N'-diacetyltetrahydro-4,4'-dipyridyl and the magnetic susceptibility (*K*) calculated in units per gram.

**Quantitative Catalytic Hydrogenation.**—Eight and six-tenths grams (0.035 mole) of yellow N,N'-diacetyltetrahydro-4,4'-dipyridyl dissolved in 130 ml. of glacial acetic acid was hydrogenated over 0.2 g. of platinum oxide at room temperature and 50 pounds per square inch initial pressure. Hydrogen was absorbed as shown in Fig. 1. Removal of the solvent under reduced pressure gave a white residue which yielded on recrystallization from dioxane 6.3 g. (74%) of shiny colorless plates of N,N'-diacetyl-4,4'-dipiperidyl, m. p. 173.5-174.5° (reported,<sup>13</sup> 174°). Attempts to hydrogenate N,N'-diacetyltetrahydro-4,4'-dipyridyl using Raney nickel in dioxane or methanol under pressures of 50-1400 pounds per square inch and temperatures of 25-70° generally failed to reduce the compound. Zinc and methanolic sodium hydroxide also failed to effect reduction.

**Absorption Spectra.**—Infrared determinations were kindly carried out by Mrs. J. L. Johnson using the crystalline white and yellow forms in Nujol. The instrument was a Perkin-Elmer Model 12B infrared spectrometer with rock salt optics.

Ultraviolet determinations were kindly made by Mr.

(12) Driggs and Hopkins, *THIS JOURNAL*, **47**, 363 (1925).

(13) Emmert and Wolpert, *Ber.*, **74**, 1015 (1941).

John C. Brantley using a Model D Beckman spectrophotometer with 95% ethanolic solutions of the white (5.22 mg. per liter of solution) and yellow (12.1 mg. per liter of solution) forms.

**Ozonolysis.**—Five and five-tenths grams (0.025 mole) of yellow N,N'-diacetyltetrahydro-4,4'-dipyridyl dissolved in 40 ml. of glacial acetic acid was ozonized for forty-eight hours at 20° with 3% ozone flowing at a rate of 2.2 ml. per minute. The acetic acid solution was then added dropwise at 0° to 300 ml. of water and 30 ml. of 30% hydrogen peroxide. The mixture was allowed to stand overnight, then heated to 90° for ten minutes. The solvent was removed by distillation, leaving a residue of 3.5 g. of crude yellowish succinic acid, m. p. 160-178°. From this was prepared a *p*-bromophenacyl ester by the method of Shriner and Fuson,<sup>14</sup> m. p. 210-212°. Succinamide was also prepared in 43% yield through the methyl ester from 0.692 g. of the crude acid, using the method of Morrell.<sup>15</sup> The colorless needles, recrystallized from water, melted at 260-261°. A mixed m. p. with an authentic sample was not depressed.

**Effect on Polymerization of Styrene.**—The flow times of solutions of 0.10 g. of the white and yellow forms of N,N'-diacetyltetrahydro-4,4'-dipyridyl in 13.3 ml. of freshly-distilled styrene were periodically compared with those of pure styrene in Ford-type viscosimeters<sup>16</sup> maintained at 58°. The viscosity of the pure styrene increased steadily, until after two days the liquid was too thick to flow. The flow times of the solutions were unchanged after two days at 58°, and still unchanged after five months at room temperature.

### Summary

Evidence is presented to suggest that N,N'-diacetyltetrahydro-4,4'-dipyridyl dissociates into free radicals which give rise to the yellow modification of the compound.

(14) Shriner and Fuson, "Identification of Organic Compounds," 2nd ed., John Wiley and Sons, Inc., New York, N. Y., 1940, p. 132.

(15) Morrell, *J. Chem. Soc.*, **105**, 2698 (1914).

(16) Foord, *ibid.*, **48** (1940).

URBANA, ILLINOIS

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[CONTRIBUTION FROM THE CHEMICAL LABORATORY OF INDIANA UNIVERSITY]

## The Reaction of Azlactones with Secondary Amines

BY DAVID K. BARNES,<sup>1</sup> E. CAMPAIGNE AND R. L. SHRINER<sup>2</sup>

No systematic study has been made of the reaction of secondary amines with azlactones.<sup>3</sup> Only a few scattered examples of the reaction have been reported in the literature.<sup>4-8</sup> In view of this fact, it was considered desirable to investigate the reactions of 2-phenyl-4-benzal-5-oxazolone (an un-

saturated azlactone) and 2-phenyl-4-benzyl-5-oxazolone (a saturated azlactone) with the following series of secondary amines: piperidine, morpholine, dimethylamine, diethylamine, methylaniline, ethylaniline, diphenylamine, indole and carbazole. These amines were chosen because they represented different degrees of basicity.

Erlenmeyer<sup>4</sup> reported that 2-phenyl-4-benzal-5-oxazolone (I) reacted with piperidine to produce  $\alpha$ -benzoylaminocinnamapiperidide with a melting point of 178°. In the present work, it has been found that two isomeric products may be isolated from the reaction of 2-phenyl-4-benzal-5-oxazolone<sup>9</sup> with piperidine. When equivalent amounts of the reactants were employed, Piperidide A,

(9) Two isomeric forms, (*cis* and *trans*) of this azlactone have been described by Carter and Risser, *J. Biol. Chem.*, **139**, 255 (1941). Only the readily available higher melting isomer was used by Erlenmeyer and also in the present work.

(1) Taken from part of a thesis submitted by David K. Barnes to the Faculty of the Graduate School in partial fulfillment of the requirements for the Degree, Doctor of Philosophy, in the Department of Chemistry, Indiana University. Present address, Standard Oil and Gas Co., Tulsa, Oklahoma.

(2) Present address: Chemistry Department, University of Iowa, Iowa City, Iowa.

(3) Carter, "Organic Reactions," Vol. III, John Wiley and Sons, Inc., New York, 1946, p. 198.

(4) Erlenmeyer, *Ber.*, **33**, 3035 (1900).

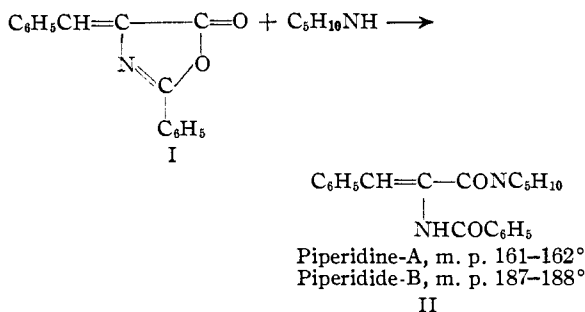
(5) Erlenmeyer and Wittenberg, *Ann.*, **337**, 294 (1904).

(6) Erlenmeyer and Stadlin, *ibid.*, **337**, 283 (1904).

(7) Lettne and Fernholz, *Z. physiol. Chem.*, **266**, 37 (1940).

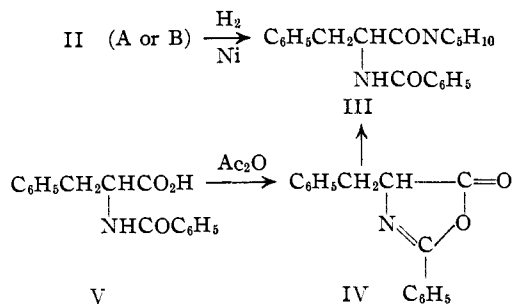
(8) Doherty, Tietzman and Bergmann, *J. Biol. Chem.*, **147**, 617 (1943).

melting at 161–162.5° was obtained. The use of a large excess of piperidine in the reaction resulted in the formation and isolation of Piperidide B, melting at 187–188°.



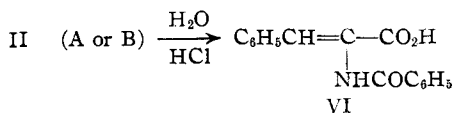
Analyses showed that both of these compounds were formed from one mole of the azlactone and one mole of piperidine. No compound corresponding to the addition of a second mole of amine to the unsaturated amide was found in spite of changes in mole ratio of reactants and time, temperature and solvents.

A study of the reactions of these isomers showed that they possessed the same chemical properties. For example, catalytic reduction of both piperidides (II, A and B) produced  $\alpha$ -benzoylamino-cinnamapiperidide (III) which was identical with a sample of this compound obtained by treatment of 2-phenyl-4-benzyl-5-oxazolone (IV) with pi-



peridine. The saturated azlactone (IV) was made by the action of acetic anhydride on *dl*-benzoyl-phenylalanine (V).

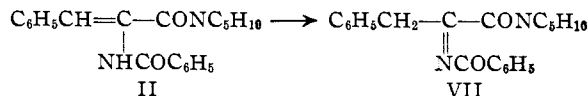
Hydrolysis of both piperidides with dilute hydrochloric acid gave piperidine and the same  $\alpha$ -benzoylamino-cinnamic acid (VI) m. p. 229–230°, which is presumably the *trans*-isomer. This was



somewhat surprising since, if the two piperidides are *cis-trans* isomers, it might be expected that the *cis*- and *trans*-forms of VI would be formed. It is of course possible that interconversion occurred during the hydrolysis.

Both piperidides were converted to the original oxazolone (I) and piperidine when heated in tetralin or glacial acetic acid. Although the isomerism

of the two compounds might be due to the shift of the double bond and proton (Formulas II, VII), the above regeneration of the oxazolone, and hy-



drolysis to VI favor the possibility that they are *cis* and *trans* isomers. It was also found that the lower melting isomer (IIA) was converted to the higher melting form (IIB) by heating with piperidine or pyridine; reagents which have previously been shown to cause conversion of *cis* to *trans* isomers.<sup>9,10</sup>

Similarly, two isomeric products were obtained from the reaction of morpholine with 2-phenyl-4-benzyl-5-oxazolone. Catalytic reduction of each isomer resulted in the formation of  $\alpha$ -benzoylamino-hydrocinnamamorpholide, and acid hydrolysis yielded  $\alpha$ -benzoylamino-cinnamic acid (VI) and morpholine.

Isomeric products were not obtained with the other amines, although there may have been small amounts of the lower melting forms present. Table I in the Experimental Part summarizes the products obtained from secondary amines and 2-phenyl-4-benzyl-5-oxazolone, and Table II, the substituted amides obtained from 2-phenyl-4-benzyl-5-oxazolone. The weakly basic amines, diphenylamine, indole and carbazole failed to open the azlactone ring.

## Experimental

**2-Phenyl-4-benzyl-5-oxazolone.**—The 2-phenyl-4-benzyl-5-oxazolone used in this work was prepared according to the method of Gillespie and Snyder.<sup>11</sup> The product was obtained in 53–73% yield, melting at 165–166° without recrystallization.

**N-Benzoyl-*dl*-phenylalanine.**—Reduction of 2-phenyl-4-benzyl-5-oxazolone with phosphorus and hydriodic acid, according to the method of Gillespie and Snyder<sup>11</sup> afforded *dl*-phenylalanine in 50–60% yields. Treatment of this product with benzoyl chloride and sodium hydroxide<sup>12</sup> resulted in the preparation of *N*-benzoyl-*dl*-phenylalanine melting at 184–186° in 70–75% yields.

**2-Phenyl-4-benzyl-5-oxazolone.**—*N*-Benzoyl-*dl*-phenylalanine was converted to 2-phenyl-4-benzyl-5-oxazolone by dehydration in acetic anhydride, according to the method of Mohr and Stroschein.<sup>13</sup> The yields ranged from 58–63% of the azlactone which melted at 70–71°. Due to the ease of hydrolysis of the saturated azlactone, the pure material was stored in a vacuum desiccator over phosphorus pentoxide prior to use.

**The Reaction of Piperidine with 2-Phenyl-4-benzyl-5-oxazolone.**—In a 25-ml. Erlenmeyer flask, 10 ml. (0.1 mole) of piperidine was added to 1.0 g. (0.004 mole) of 2-phenyl-4-benzyl-5-oxazolone. Heat was evolved as the solid azlactone dissolved. The reaction mixture was heated in a boiling water-bath for ten minutes. After cooling, the white, crystalline precipitate was filtered with suction and washed on the filter with petroleum ether. The  $\alpha$ -benzoylamino-hydrocinnamapiperidide was

(10) Lutz and Bailey, *THIS JOURNAL*, **67**, 2229 (1945); Clemons and Graham, *J. Chem. Soc.*, 213 (1930); Pollard, Bain and Adelson, *THIS JOURNAL*, **57**, 199 (1935).

(11) Gillespie and Snyder, "Org. Syntheses," Coll. Vol. II, 1943, p. 489.

(12) Steiger, *J. Org. Chem.*, **9**, 396 (1944).

(13) Mohr and Stroschein, *Ber.*, **42**, 2521 (1909).

recrystallized from dilute ethanol; the weight, after drying, was 1.3 g. (98%) of white crystals which melted at 162–162.5°.

The same procedure was used for the reaction of morpholine, methylaniline and ethylaniline with 2-phenyl-4-benzyl-5-oxazolone. Dimethylamine, diethylamine and di-*n*-butylamine gave satisfactory yields when the reaction was carried out in dry solvents such as ligroin or chloroform. The melting points and analyses of the products are given in Table I.

TABLE I

N,N-SUBSTITUTED $\alpha$ -BENZOYLAMINOHYDROCINNAMAMIDES				
Amide group	M. p. of product, °C.	Formula	Nitrogen, %	
			Calcd.	Found
Piperidide	162–162.5	C <sub>21</sub> H <sub>24</sub> O <sub>2</sub> N <sub>2</sub>	8.33	8.21
Morpholide	171–172	C <sub>20</sub> H <sub>22</sub> O <sub>3</sub> N <sub>2</sub>	8.28	8.34
Dimethylamide	148.5–149	C <sub>18</sub> H <sub>20</sub> O <sub>2</sub> N <sub>2</sub>	9.46	9.27
Diethylamide	121.5–122.5	C <sub>20</sub> H <sub>24</sub> O <sub>2</sub> N <sub>2</sub>	8.64	8.57
Methylanilide	163–164	C <sub>23</sub> H <sub>22</sub> O <sub>2</sub> N <sub>2</sub>	7.82	7.59
Ethylanilide	184.5–185	C <sub>24</sub> H <sub>24</sub> O <sub>2</sub> N <sub>2</sub>	7.52	7.33

Diphenylamine, indole and carbazole did not form substituted cinnamides. A by-product melting at 269° was formed in the reaction of the aliphatic amines with the saturated azlactone. This may be a condensation product of the azlactone with itself (see ref. 3).

**The Reaction of Piperidine with 2-Phenyl-4-benzal-5-oxazolone.** Method A.—In a 125-ml. Erlenmeyer flask fitted with a reflux condenser, 12.5 g. (0.05 mole) of 2-phenyl-4-benzal-5-oxazolone was suspended in 50 ml. of dry benzene, and 4.25 g. (0.05 mole) of freshly distilled piperidine was added. The reaction mixture warmed spontaneously, and was further heated for thirty minutes in a boiling water-bath. During this time the reaction mixture became homogeneous. Approximately two-thirds of the solvent was removed under reduced pressure, the product precipitating when the concentrated solution was allowed to cool. The white, powdery solid was filtered with suction and washed on the filter with petroleum ether. The weight of crude Piperidide A was 14.6 g. (86.5%) m. p. 151–153°. After recrystallization from 300 ml. of 95% ethanol, the product weighed 13.9 g. (83%) m. p. 161–162.5°. This melting point is the same as that of  $\alpha$ -benzoylaminohydrocinnamapiperidide but the melting point of a mixture of the two was markedly lowered.

This method was utilized for the preparation of the corresponding Morpholide A from the reaction of 2-phenyl-4-benzal-5-oxazolone with an equivalent amount of morpholine.

**Method B.**—To 5.0 g. (0.02 mole) of 2-phenyl-4-benzal-5-oxazolone was added 25 ml. (0.25 mole) of piperidine. The azlactone dissolved rapidly with the evolution of heat. The homogeneous, yellow solution was heated in a boiling water-bath for ten minutes, during which time a white solid began to precipitate. The reaction mixture was allowed to cool, and the product was filtered with suction and washed with cold acetone. The weight of crude Piperidide B was 5.0 g., m. p. 181–183°. After recrystallization from benzene, the weight was 4.2 g. (83%), m. p. 187–188°.

This method was utilized for the preparation of Morpholide B from the reaction of 2-phenyl-4-benzal-5-oxazolone with morpholine. These two general methods were used in the reaction of dimethylamine, diethylamine, methylaniline, and ethylaniline, with 2-phenyl-4-benzal-5-oxazolone. Isomers were not obtained from these amines but may have been present. Examination of the mother liquors from the crystallization of the reaction products gave small amounts of unchanged azlactone but no definite pure isomers could be separated. The melting points and analyses of the products are given in Table II.

Diphenylamine, indole and carbazole failed to react with the unsaturated azlactone. The reactants were recovered unchanged except when Method A was employed with carbazole. In this case, the recovered azlactone

TABLE II

N,N-SUBSTITUTED  $\alpha$ -BENZOYLAMINOCINNAMAMIDES

Amide group	M. p. of product, °C.	Formula	Analyses, %	
			Calcd.	Found
Piperidide (A)	161–162.5	C <sub>21</sub> H <sub>22</sub> O <sub>2</sub> N <sub>2</sub>	N, 8.38	8.43
			C, 75.45	76.14
			H, 6.56	6.80
Piperidide (B)	187–188	C <sub>21</sub> H <sub>22</sub> O <sub>2</sub> N <sub>2</sub>	N, 8.38	8.19
			C, 75.45	75.63
			H, 6.56	6.77
Morpholide (A)	135.5–137	C <sub>20</sub> H <sub>20</sub> O <sub>3</sub> N <sub>2</sub>	N, 8.33	8.17
			C, 71.43	71.65
			H, 5.95	6.06
Morpholide (B)	180.5–181	C <sub>20</sub> H <sub>20</sub> O <sub>3</sub> N <sub>2</sub>	N, 8.33	8.26
			C, 71.43	71.14
			H, 5.95	5.80
Dimethylamide	166.5–167	C <sub>18</sub> H <sub>18</sub> O <sub>2</sub> N <sub>2</sub>	N, 9.52	9.43
Diethylamide	172–173	C <sub>20</sub> H <sub>22</sub> O <sub>2</sub> N <sub>2</sub>	N, 8.70	8.70
Methylanilide	193–194	C <sub>23</sub> H <sub>20</sub> O <sub>2</sub> N <sub>2</sub>	N, 7.87	7.29
Ethylanilide	174–175	C <sub>24</sub> H <sub>22</sub> O <sub>2</sub> N <sub>2</sub>	N, 7.57	7.39

melted at 148–149°. When it was treated with pyridine it changed to the higher melting isomer, m. p. 165–166°. This corresponds to the isomerization reported by Carter and Risser.<sup>9</sup>

**Reduction of N,N-Substituted  $\alpha$ -Benzoylaminocinnamamides.**—In a 500-ml. hydrogenation bottle were placed 0.005 mole of the N,N-substituted  $\alpha$ -benzoylaminocinnamamide, 100 ml. of absolute ethanol and 2–3 g. of Raney nickel.<sup>14</sup> The bottle was placed on a Parr hydrogenation apparatus and the solution shaken with hydrogen at a pressure of 45 lb. The calculated pressure drop occurred in about five minutes, but the mixture was allowed to shake for fifteen minutes to ensure complete reduction. After removing the Raney nickel by filtration, the filtrate was concentrated to approximately one-fourth the original volume under reduced pressure. Upon cooling the solution to room temperature, a white, crystalline product precipitated which was filtered with suction and dried. The reduced products were identified by comparison with the corresponding hydrocinnamamides prepared from the reaction of 2-phenyl-4-benzyl-5-oxazolone with secondary amines (see Table I).

**Acid Hydrolysis of N,N-Substituted  $\alpha$ -Benzoylaminocinnamamides.**—The following method was used for the hydrolysis of each of the isomeric piperidides and morpholides. In a 200-ml. round-bottomed flask fitted with a reflux condenser, 0.003 mole of the N-substituted  $\alpha$ -benzoylaminocinnamamide was boiled in 100 ml. of dilute (3:1) hydrochloric acid for three hours. The white, crystalline, acid-insoluble material was filtered with suction and washed on the filter with water. When dry, this product melted at 228–230° and was readily soluble in 10% sodium hydroxide. A mixed melting point with  $\alpha$ -benzoylaminocinnamic acid showed no depression.

The acidic filtrate was cooled in an ice-water-bath, and made alkaline to litmus by the addition of 30% sodium hydroxide. A strong amine-like odor was usually noticeable. Five milliliters of benzenesulfonyl chloride and 5 ml. of 30% sodium hydroxide were added to the reaction mixture; the flask was corked tightly and shaken vigorously for thirty minutes. The white, flocculent sulfonamide was filtered with suction and washed thoroughly with water. The benzene sulfonamides were recrystallized from dilute ethanol and compared with authentic samples.

**Conversion of Piperidide A to Piperidide B.**—In a 50-ml. round-bottomed flask fitted with a reflux condenser were placed 2.5 g. (0.006 mole) of  $\alpha$ -benzoylaminocinnamapiperidide (Piperidide A), 10 ml. (0.1 mole) of piperidine and 10 ml. of benzene. The flask was immersed in a boiling water-bath and the mixture refluxed for thirty minutes. The solution became homogeneous, but after cooling, a white, crystalline precipitate was obtained. The solid was filtered with suction and washed on the filter with several small portions of petroleum ether.

(14) R. Mozingo, "Org. Syntheses," **21**, 15 (1941).

The dry material weighed 1.2 g. and melted at 166–167°. An additional 1.0 g. was obtained by evaporating the filtrate to dryness under reduced pressure. The two fractions were combined and recrystallized from benzene. The yield of pure Piperidide B was 2.0 g., melting at 187–188°. Boiling pyridine also converted the low melting isomer to the higher melting compound. In a similar way the low melting Morpholide A was converted to the higher melting Morpholide B.

### Summary

1. A series of  $\alpha$ -benzoylamino-N,N-disubstituted hydrocinnamamides has been prepared by the reaction of 2-phenyl-4-benzyl-5-oxazolone with dimethylamine, diethylamine, methylaniline, ethylaniline, piperidine and morpholine.

2. A series of  $\alpha$ -benzoylamino-N,N-disubstituted cinnamamides has been prepared by the re-

action of 2-phenyl-4-benzal-5-oxazolone with the same secondary amines. Each of these compounds was catalytically hydrogenated to the corresponding hydrocinnamamide.

3. Piperidine and morpholine were found to react with 2-phenyl-4-benzal-5-oxazolone to yield two isomeric products in each case. The isomeric piperidides were both hydrolyzed to the same  $\alpha$ -benzoylamino-cinnamic acid, and reduced to  $\alpha$ -benzoylamino-hydrocinnamapiperidide and morpholide, respectively.

4. Diphenylamine, indole and carbazole failed to react with 2-phenyl-4-benzyl-5-oxazolone or 2-phenyl-4-benzal-5-oxazolone under the conditions employed in this work.

BLOOMINGTON, INDIANA  
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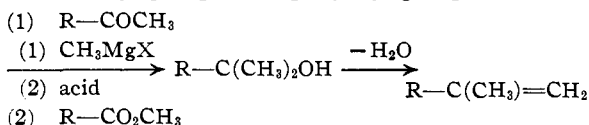
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[CONTRIBUTION FROM THE PURDUE RESEARCH FOUNDATION AND THE DEPARTMENT OF CHEMISTRY, PURDUE UNIVERSITY]

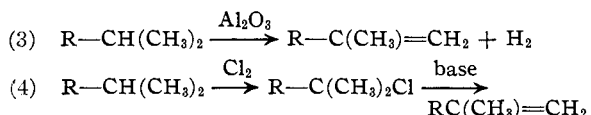
## Monomers and Polymers. III. A New Synthesis for $\alpha$ -Methylstyrenes<sup>1,2</sup>

BY G. BRYANT BACHMAN AND HENRY M. HELLMAN

Most  $\alpha$ -methylstyrenes so far described have been prepared by two general types of syntheses. The first involves the conversion of carbonyl or carbalkoxy groups to isopropenyl groups, *e. g.*



The method is limited to intermediates which contain no other groups affected by Grignard reagents and is impractical for large scale production. The second type involves the conversion of isopropyl groups to isopropenyl groups, *e. g.*



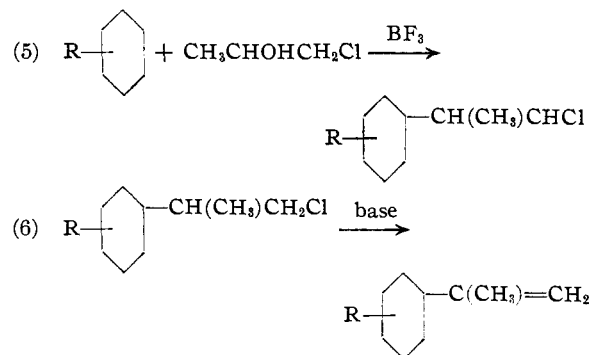
It is better suited for commercial production but is also limited as to other groups which may be present. Thus alkyl substituents are especially troublesome because of their indiscriminate attack by the dehydrogenation catalyst or by the halogen. Furthermore, the tertiary halides produced (Equation 4) tend to dehydrohalogenate during distillation giving difficulty separable mixtures.

We have sought for and found a synthesis which can be applied more or less generally to substitute  $\alpha$ -methylstyrenes, especially of the types difficultly obtainable by previously known methods,

(1) From the Ph.D. thesis of H. M. Hellman, Purdue University, June, 1947. Present address: Department of Chemistry, New York University, New York.

(2) For previous papers in this series see THIS JOURNAL, **69**, 2022 (1947); **70**, 622 (1948), and others to be published.

and which might be adapted to large scale production. The synthesis is illustrated by the following equations in which R represents one or more nuclear substituents.



Aromatic compounds have been alkylated before with alcohols and with alkyl halides<sup>3</sup> but never apparently with halohydrins. We have found that secondary alcohols react so much more readily than primary halides in this synthesis that condensation with two aryl nuclei to form diarylpropanes may be made of minor importance. Positional isomers are formed but the para derivative (with monosubstituted benzenes) is the chief product.

In Table I are shown the haloalkylation products of a number of substituted benzenes. The method appears to work especially well with alkylated benzenes, probably because the alkyl group activates the nucleus to further substitution.

(3) For a general review see C. C. Price, "The Alkylation of Aromatic Compounds by the Friedel-Crafts Method," in "Organic Reactions," John Wiley and Sons, Inc., New York, N. Y., 1946, Vol. III, Chapter 1.