of vanadium pentoxide was omitted were consistently low in carbon by 0.77–0.81%, and indicated incomplete combustion.

Hydrolysis of 0.35 g. of XVIII in 20 ml. of glacial acetic acid and 3 drops of concentrated hydrochloric acid, with refluxing for 30 min., gave 0.18 g. (65%) of IXb, m.p. 72–73°, which was identified by recrystallization from hexane (m.p. 73–74°) and mixture m.p., and by identity of infrared spectrum with that of an authentic sample.

Hydrolysis of 0.53 g. of XVIII in 8 ml. of 95% ethanol containing 0.07 g. of dissolved sodium (24 hr. at room temperature) gave 0.38 g. (75%) of crude XIV which on two recrystallizations from benzene was identified by m.p. 168–168.5°, mixture melting point, and infrared absorption spectrum identical with that of an authentic sample.

Determination of acidities (pK_a').[2b] Because of difficult solubilities in water the determinations were made in 50% ethanol at 25° and they are therefore relative only. The true acids (*trans*), and the *cis* acids, which are partly or largely in the γ-hydroxylactone forms, and the γ-anilinolactone XIV, were sufficiently acidic so that the relative pK_a' values could be calculated from the apparent *p*H values at half neutralization as determined by means of a Beckman Model G *p*H meter. For calculation of the relative pK_a' values for the very weak acids, the γ-hydroxylactams, the apparent *p*H values were determined by the use of trinitrotoluene as an indicator for the range involved and a series of standard sodium hydroxide solutions, and by measuring absorptivities at 450 mμ, with time standardization to allow for slow deterioration of the standards. The relative pK_a' values were: Xa (*cis*), 6.39: IXa (*trans*), 4.37; Xc, 8.69; IVa, 11.8; IVb, 11.7; XIV, 8.40.

CHARLOTTESVILLE, VA.

[CONTRIBUTION FROM THE PIONEERING RESEARCH DIVISION, TEXTILE FIBERS DEPARTMENT, E. I. DU PONT DE NEMOURS & COMPANY, INC.]

Some Reactions of *p*-Toluenesulfonyl Isocyanate

C. KING

Received October 12, 1959

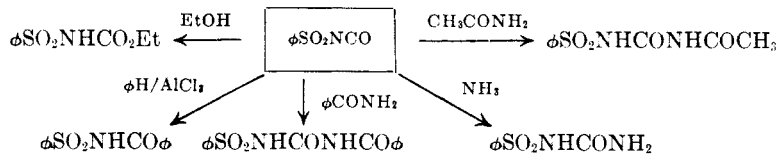
Several new reactions of *p*-toluenesulfonyl isocyanate (I) are reported. Reaction with *N,N*-dialkylamides gives *N,N*-dialkyl-*N'*-*p*-toluenesulfonylamidines (II), with elimination of carbon dioxide. Monoalkylamides give rise to *N*-alkyl-*N*-acyl-*N'*-*p*-toluenesulfonylureas (III) by simple addition, or to *N*-alkyl-*N'*-*p*-toluenesulfonylamidines (IV), with elimination of carbon dioxide. Isobutyraldehyde reacts to give VI. Dimethyl sulfoxide yields *N*-*p*-toluenesulfonyl dimethyl sulfilimine (VII).

Recent published work[1] has described reactions of *p*-toluenesulfonyl isocyanate (I), with monoalkyl- and dialkylamides. We wish to report work done independently in this laboratory on these and other unusual reactions of *p*-toluenesulfonyl isocyanate.

Sulfonyl isocyanates have only recently become readily and economically accessible from high temperature phosgenation of sulfonamides in inert solvents.[2] Billeter[3] had prepared methane- and benzenesulfonyl isocyanates by treating the appropriate sulfonyl chlorides with silver cyanate at 120–140°. With benzenesulfonyl isocyanate he observed typical addition reactions of isocyanates. He further noted that sulfonyl isocyanates undergo ready hydrolysis to sulfonamides, rather than to ureas, which are the usual hydolysis products of isocyanates. Prior to the work of Logemann this appears to be the only reported work on sulfonyl isocyanates.

The work in this paper deals with reactions on *p*-toluenesulfonyl isocyanate, for which a convenient preparation has been described in the patent literature.[2]

Reaction with N,N-dialkylamides. Smooth, rapid reaction occurred when *p*-toluenesulfonyl isocyanate was added to an excess of *N,N*-dialkylamide at room temperature. Reactions were exothermic, and it was not found necessary to add solvent or catalyst. Good yields of crystalline solids were obtained which were products of intermolecular elimination of carbon dioxide. Dialkylamides found to react in this manner were dimethylformamide, dimethylacetamide, *N*-formylpiperidine, *N*-acetylpiperidine, and *N*-methylpyrrolidone. It is believed that the reaction proceeds through a cyclic intermediate, leading to *N,N*-dialkyl-*N'*-*p*-toluenesulfonylamidines. Thus, for the reaction with dimethylformamide:



(1) W. Logemann, D. Artini, G. Tosolini, and F. Piccini, *Ber.* **90**, 2527 (1957); **91**, 951, 2566 (1958).

(2) British Patent **692,360**, June 3, 1953.

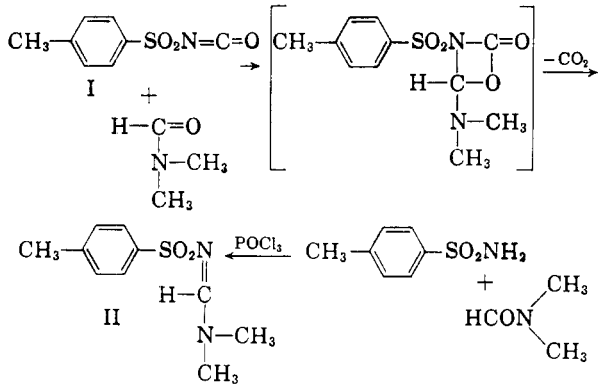
(3) O. C. Billeter, *Ber.*, **36**, 690–6 (1904); **37**, 2013–15 (1905).

TABLE I

N,N-DIALKYL-*N'*-*p*-TOLUENESULFONYL AMIDINES

Sulfonylamidine	M.P.	Yield, %	Analyses	Infrared
				—C=N—
TosN=C(H)—N(CH₃)₂	133–133.5[a]	67	Calcd. for $C_{10}H_{14}SO_2N_2$: C, 53.07; H, 6.25; N, 12.38; S, 14.25.	Absorption
			Found: C, 53.2; H, 6.0; N, 11.9; S, 14.0.	1630 cm.$^{-1}$ 6.11 μ
TosN=C(CH₃)—N(CH₃)₂	120.5–121[a]	95	Calcd. for $C_{11}H_{16}SO_2N_2$: C, 54.95; H, 6.72; N, 11.66; S, 13.33.	
			Found: C, 55.0; H, 6.53; N, 11.35; S, 13.29.	1580 cm.$^{-1}$ 6.31 μ
TosN=C(H)—N(C₅H₁₀)	147.5–149[a]	46	Calcd. for $C_{13}H_{18}SO_2N_2$: C, 58.61; H, 6.82; N, 10.32; S, 12.03.	
			Found: C, 57.9; H, 6.8; N, 10.2; S, 12.1.	1540 cm.$^{-1}$ 6.5 μ
TosN=C(CH₃)—N(C₅H₁₀)	145–147	64	Calcd. for $C_{14}H_{20}SO_2N_2$: C, 59.96; H, 7.20; N, 9.99; S, 11.43.	
			Found: C, 59.1; H, 7.0; N, 9.8; S, 11.6.	1610 cm.$^{-1}$ 6.2 μ
TosN=C(N(CH₃)—CH=CH—CH=CH—)	163–164	32	Calcd. for $C_{12}H_{16}SO_2N_2$: C, 57.12; H, 6.39; N, 11.10; S, 12.71.	
			Found: C, 57.1; H, 6.3; N, 11.1; S, 12.8.	1600 cm.$^{-1}$ 6.25 μ

[a] Logemann reports 133–134°, 121–124°, and 146–148° for these compounds, respectively.



Structural assignment of II was based on elemental and infrared analysis, and on establishing its identity by mixture melting point with *N,N*-dimethyl-*N'*-*p*-toluenesulfonyl formamidine prepared from condensation of *p*-toluenesulfonamide with dimethylformamide. This latter preparation was adapted from a recently disclosed[4] method.

The cyclic mechanism postulated above for sulfonylamidine formation seems plausible, particularly in view of the high polarity of the reactants. The isocyanate function is rendered highly electrophilic by the adjacent sulfonyl groups, and would be highly attracted to the electron-rich carbonyl of a dialkylamide.

Analogous amidine formation is known for ordinary isocyanates. Thus, treatment of benzanilide with phenyl isocyanate at 200–220° gives *N,N'*-diphenylbenzamidine.[5]

Table I summarizes the data on reaction products from dialkylamides.

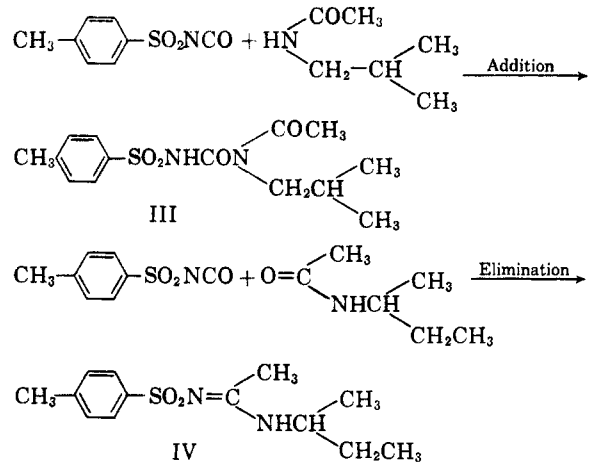
Reaction with unsubstituted amides. Acetamide and formamide gave products of simple addition, *N*-toluenesulfonyl-*N'*-acylureas. A second product, however, was isolated from the reaction with formamide. It melted at 195–196°, and had the elemental composition of *N*-*p*-toluenesulfonyl urea. This latter product arose apparently from hydrolysis during work up of the primary product, *N*-*p*-toluenesulfonyl-*N'*-formyl urea. *N*-*p*-toluenesulfonyl urea is reported to melt at 192°.[6]

No evidence was found for reaction at the carbonyl group, as in the case of the dialkylamides.

Reaction with monosubstituted amides. Reaction of *p*-toluenesulfonyl isocyanate with *mono*alkylamides was not so clean as those with unsubstituted or dialkylamides. Crystalline products were obtained only as minor constituents, while for the most part reaction mixtures were noncrystallizable, frequently dark, oils. Reaction mixtures involving formamides occasionally had a carbylamine-like odor, arising presumably from dehydration of the formamides.

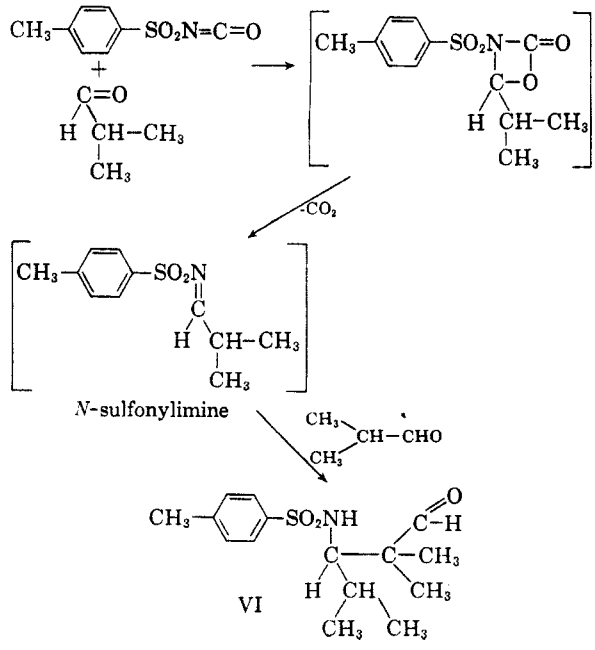
Crystalline compounds were isolated, however, which were products of simple addition, as for the unsubstituted amides, and products formed by elimination of carbon dioxide, as for the dialkylamides. Thus, *N*-isobutylacetamide reacted by addition, and *N*-*sec*-butylacetamide reacted by elimination. These products were identified by infrared and elemental analysis.

(4) German Patent **949,285**, Sept. 20, 1956.
(5) P. F. Wiley, *J. Am. Chem. Soc.*, **81**, 3746 (1949).
(6) G. H. Cox and S. M. Raymond, Jr., *J. Am. Chem. Soc.*, **63**, 300–301 (1941).



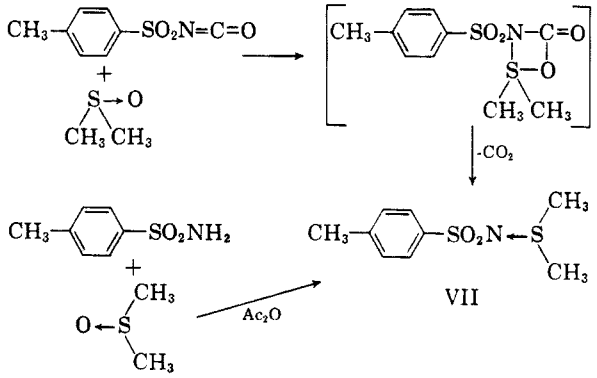
The crystalline product obtained from *N*-isobutylformamide was *N*-*p*-toluenesulfonyl-*N'*-isobutyl urea, (V), identified by infrared and elemental analysis, and by comparison with authentic urea prepared by addition of isobutylamine to *p*-toluenesulfonyl isocyanate. Formation of V could have arisen from hydrolysis of a product-by-addition (analogous to III) or from hydration of a product-by-elimination (analogous to IV). In either case, the urea is a product of a secondary reaction, occurring presumably during work up.

Reaction with aldehydes and ketones. Isobutyraldehyde reacted moderately fast to give a somewhat unstable crystalline solid, m.p. 147.5–148°, having the composition $C_{15}H_{23}SO_3N$. This composition indicates that two molecules of isobutyraldehyde reacted with one of *p*-toluenesulfonyl isocyanate, with elimination of carbon dioxide. The reaction may be formulated as sulfonylimine formation followed by addition of isobutyraldehyde. Infrared analysis disclosed the presence of —NH and of >C=O, which had the appearance of a hydrogen-bonded carbonyl. The carbonyl group might well be expected to bond with the sulfonamide hydrogen. An attempt to prepare a sulfonylimine with *p*-methylbenzaldehyde (no α-hydrogen for further addition) failed, presumably because of hydrolysis. Reaction with this aldehyde gave solid which initially melted at 90–93° but which, on attempted purification, yielded only *p*-toluenesulfonamide contaminated with aldehyde.



The ready reaction of *p*-toluenesulfonyl isocyanate with amides and aldehydes suggested that reaction might occur with other carbonyl compounds. No reaction was observed, however, with acetone, cyclohexanone, benzophenone, acetyl chloride, or ethyl acetate.

Reaction with sulfoxides. With dimethyl sulfoxide a rapid exothermic reaction occurred, giving crystalline product in high yield. This product proved to be *N*-*p*-toluenesulfonyl dimethyl sulfilimine (VII), since it had the appropriate elemental composition and did not depress the melting point of an anthentic sample prepared by condensing *p*-toluenesulfonamide with dimethyl sulfoxide in acetic anhydride. Infrared spectra of samples from both preparations were identical. Sulfilimine formation may, formally, at least, proceed through a cyclic intermediate.



Reaction with amines and mercaptans. A number of more conventional derivatives of *p*-toluenesulfonyl isocyanate were prepared. Five amines gave the expected ureas by simple addition. Dodecyl mercaptan reacted slowly to give the thiourethane, m.p. 61–63°. Table II summarizes these results.

EXPERIMENTAL

p-Toluenesulfonyl isocyanate (I). The following procedure is substantially that of British Patent 692,360. Phosgene, 700 ml., was condensed in a 1-l. round bottomed flask and allowed to evaporate at room temperature through a stirred, refluxing solution of 1000 g. *p*-toluenesulfonamide in 3500 ml. 1,2,4-trichlorobenzene. After completion of the addition the trichlorobenzene was removed by distillation at reduced pressure through a 6-in. Vigreux column (90° at 100 mm.); distillation was continued until the head temperature was 110°. The residue was distilled at 0.05 mm., and, after a short forerun, the main portion passed over at 87–90°; a little was collected up to 107°. Fractionation of this dis-

TABLE II

PRODUCTS FROM AMINES AND MERCAPTANS

Urea	M.P.	Yield, %	Analyses
TosNHCONHϕ	169–170	83	Calcd. for $C_{14}H_{14}SO_3N_2$: C, 57.91; H, 4.87; N, 9.65; S, 11.04. Found: C, 58.0; H, 4.8; N, 9.5; S, 10.8.
TosNHCONH-*iso*-Bu	169–169.5[a]	74	Calcd. for $C_{12}H_{18}SO_3N_2$: C, 53.30; H, 6.72; N, 10.38; S, 11.86. Found: C, 53.1; H, 6.63; N, 10.3; S, 11.8.
TosNHCO-N⟨⟩	148–149	65	
TosNHCONH-*sec*-Bu	123–125[a]	81	
TosNHCONH-*n*-Bu	117–122[a]	44	
$TosNHCOSC_{12}H_{25}$	61–63	90	

[a] Logemann reports 168–170°, 124–126°, and 124–126° for these compounds, respectively.

tillate through a 12-in. glass helix packed column afforded 597 g. of pure *p*-toluenesulfonyl isocyanate, b.p. 90–93° at 0.05 mm. (54%). It was necessary to store this compound under dry nitrogen in tightly stoppered bottles, since even short exposure to atmospheric moisture caused hydrolysis to *p*-toluenesulfonamide. The boiling range has previously been recorded as 114–116° at 0.3 to 0.5 mm.[2]

N,N-Dimethyl-N'-p-toluenesulfonyl formamidine (II). A. Ten g. of I was slowly added to 15 ml. dry dimethylformamide swirled in a 50-ml. Erlenmeyer flask. Rapid heating occurred, with rapid evolution of carbon dioxide. The colorless mixture was allowed to stand at room temperature overnight, and was then poured into water to precipitate product, which was collected on a filter. Recrystallization from an ethanol-water mixture gave white crystals, m.p. 133–134°, 8.8 g. (67%). Strong absorption in the infrared at 6.11μ indicated a —C=N-grouping.

B. A solution of 10.3 g. (0.06 mole) of *p*-toluenesulfonamide, 6.0 g. (0.10 mole) dimethylformamide, and 11.6 g. (0.076 mole) phosphorus oxychloride in 40 ml. dry toluene was refluxed for 4 hr. The reaction mixture was cooled, stirred into 150 ml. ice water, and then neutralized with sodium bicarbonate. The crystals which separated were collected on a filter and then recrystallized from benzene. Product melted at 132–134°, and amounted to 3.3 g. (26%). The melting point of a mixture of product from A and B was 133–134°. The infrared spectrum of B was identical with that of A.

N-p-Toluenesulfonyl-N'-acetylurea. Acetamide, 5 g. (0.083 mole) was added with shaking to 10 g. (0.058 mole) (I). When the acetamide was nearly all dissolved a vigorous reaction occurred and the reaction mixture solidified. Crystallization from ethanol afforded 11.7 g. (79%) of white crystals, m.p. 178–179°. Infrared analysis showed —NH absorption at 3.17μ, and the split carbonyl absorption expected for the grouping —C(=O)—N(—)—C(=O)— (5.75 and 5.94 μ).

Anal. Calcd. for $C_{10}H_{12}SO_4N_2$: C, 46.86; H, 4.73; N, 10.93; S, 12.82. Found: C, 46.9; H, 4.7; N, 10.9; S, 12.9.

N-p-Toluenesulfonyl-N'-formylurea. Formamide, 15 ml., was mixed with 10 g. (0.058 mole) (I), and the mixture was allowed to stand for 4 days at room temperature. The product purified by crystallization from water amounted to 10.0 g. (57%), m.p. 155–156°. Infrared absorption at 5.72 and 5.9 μ indicated —C(=O)—N(—)—C(=O)—.

Anal. Calcd. for $C_9H_{10}SO_4N_2$: C, 44.61; H, 4.17; N, 11.56; S, 13.23. Found: C, 44.8; H, 4.3; N, 11.5; S, 13.2. Concentration of the mother liquors yielded 2.7 g. of a white solid melting at 193–197°. Crystallization from ethanol-water gave pure *N-p*-toluenesulfonyl urea, m.p. 195–196° (lit.[2] 192°).

Anal. Calcd. for $C_8H_{10}SO_3N_2$: C, 44.83; H, 4.71; N, 13.08. Found: C, 44.7; H, 4.6; N, 12.9.

N-p-Toluenesulfonyl-N'-isobutylurea (III). Ten g. of I (0.058 mole) was added to 15 ml. *N*-isobutylacetamide at room temperature. Reaction was slow, with slow evolution of gas. The mixture was allowed to stand at room temperature overnight, and was then taken up in ethanol. Addition of water with cooling in ice caused separation of crystals which were purified by recrystallization from ethanol-water. Yield was 1.7 g. (6%); m.p. 80–81°. Infrared absorption at 5.82 and 5.99 μ indicated the grouping —C(=O)—N(—)—C(=O)—.

Anal. Calcd. for $C_{14}H_{23}SO_4N_2$: C, 53.31; H, 7.36; N, 8.88; S, 10.16. Found: C, 53.77; H, 6.40; N, 8.9; S, 10.2.

N-sec-Butyl-N'-p-toluenesulfonylacetamidine (IV). Ten g. of I (0.058 mole) was mixed with 15 ml. *N-sec*-butylacetamide. A moderately vigorous reaction occurred with gas evolution to give a thick yellowish liquid. A small amount of white solid was obtained when this liquid was cooled and mixed with water. Recrystallization from ethanol-water gave 0.46 g. (2.6%) of crystalline product, m.p. 86.5–88°. Infrared absorption at 3.07 μ indicated —NH, and that at 6.47 μ indicated —C=N—. The remaining oil was not further examined.

Anal. Calcd. for $C_{13}H_{20}SO_2N_2$: C, 58.17; H, 7.53; N, 10.44; S, 11.94. Found: C, 58.23; H, 7.49; N, 10.4; S, 11.9.

N-p-Toluenesulfonyl-N'-isobutylurea. A. *N*-Isobutylformamide, 15 ml., was mixed with 10 g. (0.058 mole) of (I). A very rapid darkening occurred, with evolution of gas. The reaction mixture was a dark oil having the odor of a carbylamine. It was allowed to stand 3 days, and then stirred into cold water. A small amount of solid which separated was crystallized several times from ethanol-water. Yield was 0.36 g. of white crystals melting at 169.5–70.5°.

Anal. Calcd. for $C_{12}H_{18}SO_3N_2$: C, 53.50; H, 6.72; N, 10.36; S, 11.86. Found: C, 53.4; H, 6.3; N, 10.3; S, 11.8.

B. To 15 ml. of isobutylamine was added, in portions at room temperature, 10 g. (0.058 mole) I. The reaction was very vigorous. Product crystallized from ethanol-water melted at 169–170°, and amounted to 11.4 g. (74%). The melting point of a mixture of product from A and B melted at 169–170°.

Reaction with isobutyraldehyde (VI). Isobutyraldehyde, 15 ml., was mixed with 10 g. (0.058 mole) I at room temperature. After a short induction period the reaction mixture warmed to about 50° and evolved gas. The mixture was allowed to stand overnight, and was then taken up in methylene chloride and cooled in Dry Ice. White solid which separated was collected on a filter and washed with cold ether. When dry the product melted at 143–145°. Three recrystallizations from chloroform–petroleum ether afforded 3.9 g. (23%), m.p. 147.5–148.5°. The compound was somewhat unstable; a strong odor of isobutyraldehyde developed from purified samples that had stood for about

a week. Infrared analysis disclosed —NH absorption at 3.01 μ, and the absorption of a hydrogen-bonded carbonyl at 5.83 μ.

Anal. Calcd. for $C_{15}H_{23}SO_3N_2$: C, 60.65; H, 7.81; N, 4.71; S, 10.78. Found: C, 60.4; H, 7.6; N, 4.65; S, 10.7.

N-p-Toluenesulfonyl dimethyl sulfilimine (VII). A. Ten g. (0.058 mole) of I was added slowly to 15 g. (0.19 mole) of dry dimethyl sulfoxide with stirring. Rapid heating occurred, with evolution of gas. The colorless reaction mixture was allowed to stand at room temperature for 5 hr. The product was then isolated by precipitation with water, followed by crystallization from ethanol-water mixture. Yield of pure sulfilimine, m.p. 157–158°, was 11.7 g. (87%).

B. This preparation was adapted from the procedure of Tarbell and Weaver[7] for sulfilimine formation. A solution of 8.6 g. *p*-toluenesulfonamide (0.05 mole) and 3.9 g. (0.05 mole) dimethyl sulfoxide in 25.5 g. acetic anhydride was heated over a steam bath for 1 hr. The solution was then cooled to room temperature and stirred into an ice-cold solution of 20 g. sodium hydroxide in 60 ml. water. The white crystals which separated were collected on a filter and then recrystallized from benzene. Yield was 5.5 g. (47%), m.p. 157–158°. Infrared spectra of samples from A and B were identical.

WILMINGTON 98, DEL.

(7) D. S. Tarbell and C. Weaver, *J. Am. Chem. Soc.*, **63**, 2939 (1941).

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

Acenaphthene Chemistry. VI.[1,2] Preparation and Reactions of Some Pyracene Glycols

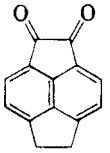
HENRY J. RICHTER AND WILLIAM C. FEIST

Received September 14, 1959

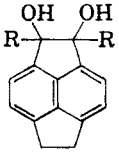
1,2-Diphenylpyracenediol reacts with 47% aqueous hydriodic acid to form the stable orange-colored 1,2-diphenyl-5,6-dihydropyracylene (III). With iodine in glacial acetic acid, the diol is converted to 2,2-diphenylpyracenone-1. The reduction of 1,2-diketopyracene with sodium borohydride produces an equimolar mixture of *cis* and *trans* 1,2-pyracenediols. The rearrangement of these diols affords pyracenone-1.

A number of derivatives of pyracene have been described in the literature.[1,3–5] There are also described some attempts to prepare the conjugated-unsaturated nonalternate hydrocarbon pyracylene.[5b] Anderson and Anderson discuss the calculated resonance energy and the 1,2- and 5,6-bond distances and conclude that such a molecule could exist. With the preparation of 1,2-diketopyracene[1] (I), the introduction of groups into the pyracene molecule has been simplified. This paper describes the preparation and reactions of a number of these derivatives.

1,2-Diketopyracene (I) was prepared as described by Richter and Stocker[1] and treated with phenylmagnesium bromide to form the *trans* pinacol II. The pinacol II reacted with 47% hydriodic acid to afford 66% of 1,2-diphenyl-5,6-dihydropyracylene (III). This bright orange compound is stable even at its melting point of 226°. The unsubstituted hydrocarbon, 1,2-dihydropyracylene, prepared by Anderson and Anderson[5b] is reported to decompose at room temperature.



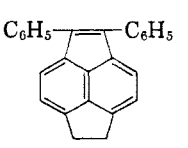
I



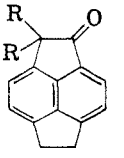
II. R = C_6H_5
Va. R = H, *cis*
Vb. R = H, *trans*

A similar elimination with 47% hydriodic acid was observed with 1,2-diphenylacenaphthenediol-1,2 which formed 1,2-diphenylacenaphthylene in good yield. The course of this elimination is uncertain as no iodine color is observed. This does not exclude the formation and decomposition of a 1,2-diiodo compound as iodine dissolves in 47% hydriodic acid to form a colorless solution.[6]

When II was treated with iodine in boiling glacial acetic acid according to Bachmann and Chu,[7] rearrangement of the pinacol led to the formation of 2,2-diphenylpyracenone-1 (IV) in 73.5% yield.



III



IV. R = C_6H_5
VI. R = H

(1) Previous paper: H. J. Richter and F. B. Stocker, *J. Org. Chem.*, **24**, 366 (1959).

(2) This work was supported by the National Institute of Health, Grant Cy-2997-C3.

(3) M. C. Kloetzel and F. L. Chub, *J. Am. Chem. Soc.*, **72**, 150 (1950).

(4) A. G. Anderson, Jr., and R. H. Wade, *J. Am. Chem. Soc.*, **74**, 2274 (1952).

(5) A. G. Anderson, Jr., and R. G. Anderson, (a) *J. Org. Chem.*, **22**, 1197 (1957). (b) *J. Org. Chem.*, **23**, 517 (1958).

(6) C. A. Jacobsen, *Encyclopedia of Chemical Reactions*, Vol. III, Reinhold Publishing Corp., New York, N. Y., 1949, p. 709.

(7) W. E. Bachmann and E. J. Chu, *J. Am. Chem. Soc.*, **58**, 1118 (1936).