The reaction of VIII *with methanolic potassium hydroxide.* To a 250 ml., three necked flask fitted with a reflux condenser, a stirrer, and a dropping funnel was added 30 g. (0.148 mole) of VIII. Potassium hydroxide (17 g., 0.30 mole) dissolved in 100 ml. of methyl alcohol was added dropwise while the reaction flask was maintained at 30–40°. After addition was complete, the reaction mixture was poured into 500 ml. of cold water. The organic layer was separated, washed with water, dried over calcium chloride, and distilled to give 8 g. (30.2% conversion) of material, b.p. 181°, n_D^{19} 1.4290, d_4^{19} 1.2654.

Anal. Calcd. for $C_7H_8F_2O_3$: MR_D, 35.63; %C, 47.19; %H, 4.53; %F, 21.33. Found: MR_D, 36.44; %C, 47.18; %H, 4.68; %F, 21.12.

NMR spectra. Spectra were obtained for solutions of 1:1 volume ratio in carbon tetrachloride, using a Varian high-resolution spectrometer operating at a frequency of 56.4 megacycles. The reference was ethyl trifluoroacetate in the external part of a precision coaxial tube. Sidebands were applied by an audio oscillator continuously monitored by a Hewlett-Packard frequency counter, and the results were averaged for several sweeps through the spectrum of each sample. A shift of resonance up-field is reported as a positive chemical shift. The maximum uncertainty of the chemical shifts is estimated to be 0.03 p.p.m.

Acknowledgment. We wish to acknowledge the financial support of this research by the Office of the Quartermaster General under Contract DA-19-129-QM-500, Dr. J. C. Montermoso as project officer. We wish to thank Dr. L. D. La Zerte of the Minnesota Mining & Manufacturing Co. for furnishing the sample of perfluoroacrylonitrile used in the experiments.

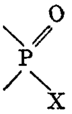
GAINESVILLE, FLA.

[CONTRIBUTION FROM THE RESEARCH LABORATORIES, ARMOUR INDUSTRIAL CHEMICAL COMPANY, McCOOK, ILL.]

A New Phosphorylation Reaction of Olefins. I. Scope of the Reaction

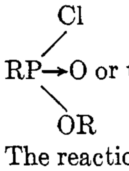
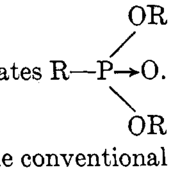
ERIC JUNGERMANN,[1] J. J. McBRIDE, JR., R. CLUTTER, AND AGO MAIS

Received July 7, 1961

A new general method for forming a phosphorus-carbon bond at an olefinic linkage is described. It is based on the reaction of an olefin with phosphorus trichloride in the presence of aluminum chloride. The resultant products are saturated phosphorus-containing acids or acid chlorides of the type  $>P(=O)X$, where X = OH or Cl. Esters can be prepared by reaction of the chlorides with alkoxides.

Among the olefins investigated are: 2,4,4-trimethyl-2-pentene; propylene trimer, tetramer, and pentamer, methyl oleate, tricosene-11, 2-pentene, 2-methyl-2-butene, 4-methyl-2-pentene, and cyclohexene. The effect of reaction conditions on the products formed is discussed in some detail. This reaction offers a new, versatile procedure for synthesizing organic phosphorus compounds from olefins.

As part of a program of finding new derivatives of fatty acids, it was decided to investigate the possibility of adding a phosphorus containing moiety to the double bond of oleic acid. Several techniques had been reported previously: Sasin *et al.*[2] and Stiles *et al.*[3] had reported the preparation of dialkyl alkyl phosphonates by the free radical addition of dialkyl phosphites to olefins, including methyl oleate, using a peroxide catalyst. The direct addition of yellow phosphorus in the presence of oxygen has also been the subject of numerous discussions in the literature.[4,5] The free radical catalyzed addition of phosphorus trichloride to *alpha*-olefins had been reported by Kharasch,[6] but yields were low, and the scope of the reaction was limited. No work has been reported in the literature on the formation of carbon-phosphorus bonds by the addition of phosphorus trichloride to olefins under ionic conditions.

About ten years ago Clay[7] and Kinnear and Perren[8] reported that alkyl halides react with phosphorus trichloride in the presence of aluminum chloride to form a complex $[RPCl_3]^+[AlCl_4]^-$. Controlled hydrolysis of the complex yields the alkylphosphonyl chloride. Hoffmann[9] extended the reaction and by a simple batch process obtained the corresponding alkyl alkylphosphonochloridates,  $RP(Cl)(OR)\rightarrow O$ or the dialkyl alkylphosphonates  $R{-}P(OR)_2\rightarrow O$. The reaction is formally related to the conventional Friedel-Craft condensation for the preparation of aromatic phosphorus halides. $ArH+PCl_3 \xrightarrow{AlCl_3}$

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[1] Present address: Research Department, Soap Division, Armour and Company, 1355 West 31st Street, Chicago, Ill.

[2] R. Sasin, Wm. F. Olszweski, J. R. Russell, and D. Swern, *J. Am. Chem. Soc.*, **81**, 6275 (1959).

[3] A. R. Stiles, W. E. Vaughn, and F. F. Rust, *J. Am. Chem. Soc.*, **80**, 714 (1958).

[4] R. Wilstaetter and E. Sonnenfeld, *Ber.*, **47**, 2801 (1914).

[5] C. Walling, F. Stacey, R. Jamison, E. Saunders, and E. S. Huyser, *J. Am. Chem. Soc.*, **80**, 4543 (1958).

$ArPCl_2 + HCl$. The attacking species, PCl_2^+, in this reaction is probably formed as follows: $PCl_3 + AlCl_3 \rightarrow PCl_2^+AlCl_4^-$. Because of the possibility that under ionic conditions the reactions of alkyl halides and olefins both might proceed *via* a carbonium intermediate, it was decided to investigate whether the electron rich olefinic double bond would react with the $PCl_2^+AlCl_4^-$ reagent to form a carbon-phosphorus bond. Such a reaction would be novel and have advantages from the point of view of economics and versatility over the known methods.

The present paper deals with the reaction of olefins with phosphorus trichloride in the presence of aluminum trichloride. In the preliminary work, methyl oleate was used as the olefinic starting material utilizing a hundred per cent excess of both phosphorus trichloride and aluminum chloride in methylene chloride as the solvent. The reaction proceeded rapidly, indicating that the concept of using the $PCl_2^+ AlCl_4^-$ complex as the phosphorylating agent had merit. A product was isolated which contained one phosphorus atom attached directly to the fatty carbon skeleton at the site of the double bond. Extension of the reaction to other materials indicated that the reaction was general for all types of olefins.[10]

The following steps are involved in the new phosphorylation procedure: (1) formation of the $PCl_3 \cdot AlCl_3$ complex in methylene chloride, (2) addition of the olefin at room temperature, (3) reaction of the reaction complex with methanol, and (4) decomposition with water and subsequent work-up of sample.

The analytical characteristics of the product obtained in high yield from methyl oleate are summarized below. Results varied slightly depending on exact reaction and work-up conditions; the range of results covering a large number of determinations is shown in Table I.

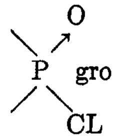
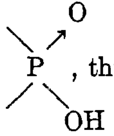
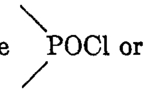
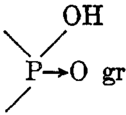
These results indicate that reaction occurred at the double bond, and that a  $\overset{O}{\underset{CL}{>P}}$ grouping was introduced, which partially hydrolyzed to the corresponding acid,  $\overset{O}{\underset{OH}{>P}}$, thus yielding a mixture of the two. Infrared absorption at 8 μ confirms presence of a —P→O group, while the resistance to hydrolysis and failure to observe P–O–C bonding in the 10-μ region of the infrared indicates that the phosphorus is bonded directly to the carbon. The loss of unsaturation was indicated by infrared analysis as well as the iodine value determination.

TABLE I

Products Obtained from Methyl Oleate

	Theoretical		
	$C_{17}H_{33}$-(POCl)-$COOCH_3$	$C_{17}H_{35}$-(POOH)-$COOCH_3$	Found (Range)
Saponification equivalent	189	180	190–200
Phosphorus	8.2	8.6	8.0 ± 0.3
Iodine value[a]	0	0	7–9
Chlorine	9.4	0	3–6

[a] Iodine value of starting methyl oleate: theoretical, 85.6; found, 88.1.

Though the exact attachment of the  $>POCl$ or  $\overset{OH}{>P}\rightarrow O$ grouping to the carbon chain was not yet established, it was decided to look into some of the reaction variables, particularly variations in steps (2) and (3). The olefin studied for this purpose was methyl oleate.

First, the effect of time and temperature on step (2) was examined. The results are shown in Fig. 1.

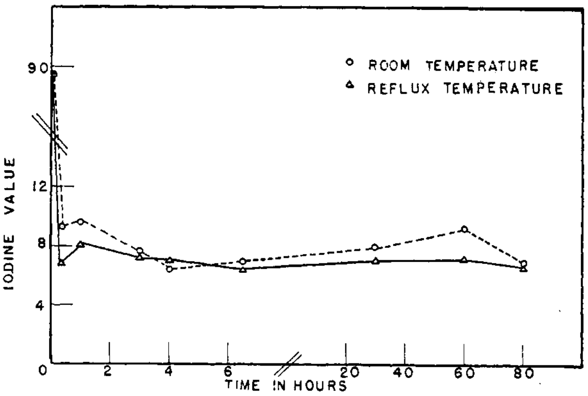


Fig. 1. Effect of temperature and time on Step (2)

Results indicate that neither time nor temperature had a great effect on the extent of reaction taking place at the double bond. The small amount of residual unsaturation (I.V. 7–9) is believed to be due to the presence of some polyunsaturation in the starting methyl oleate.

Effect of time and temperature on step (3), that is, the reaction of the methyl oleate —PCl_3-$AlCl_3$ complex with methanol, brought some interesting results. When the reaction was carried out at room temperature, time had no appreciable effect. On the other hand when the reaction mixture was refluxed

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(6) M. S. Kharasch, E. V. Jensen, and W. H. Urry, *J. Am. Chem. Soc.*, **67**, 1864 (1945).

(7) J. P. Clay, *J. Org. Chem.*, **16**, 892 (1951).

(8) A. M. Kinnear and E. A. Perren, *J. Chem. Soc.*, 3437 (1952).

(9) F. W. Hoffmann, T. C. Simmons, and L. J. Glunz, *J. Am. Chem. Soc.*, **79**, 3570 (1957).

(10) E. Jungermann and J. J. McBride, Jr., *J. Org. Chem.*, **26**, 4182 (1961).

with the methanol some significant changes occurred with time: The titratable free acid increased, the chlorine content decreased, while the saponification equivalent (total of free acid and ester) stayed constant at *circa* 190–200. These results are illustrated in Fig. 2.

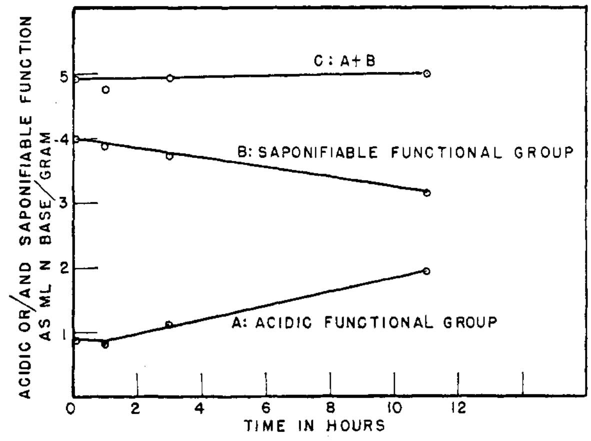


Fig. 2. Effect of time and temperature on Step (3)

The physical properties of the material isolated also varied with time, changing from a thin oil to a viscous oil to rubbery solid. Apparently, polymerization occurred accompanied by loss of chlorine and increased acidity. The phosphorylated olefins can also be polymerized by heating the finished product *in vacuo*. Other phosphorylated olefins, such as the derivatives from propylene trimer and tricosene-11 were examined and gave similar results. Further work to study the nature of the polymerization process is in progress and will be the subject of a subsequent publication.

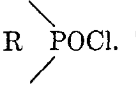
The general phosphorylation reaction was tried with a series of olefins. Commercial olefins obtained by the polymerization of propylene, such as propylene trimers (nonene), tetramers, and pentamers, all reacted readily to give in high yield primarily a phosphorus acid chloride with the general structure  $\mathrm{R}{<}\mathrm{POCl}$. These olefins are mixtures of several isomers. The typical composition of a commercial nonene is shown in Table II.

TABLE II

Isomer Distribution of Commercial Nonene[a]

$RCH{=}CH_2$	4%
$RCH{=}CHR$	15%
$R_2C{=}CH_2$	11%
$R_2C{=}CHR$	37%
$R_2C{=}CR_2$	33%

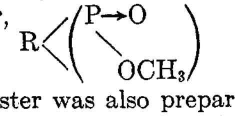
[a] Information supplied by Enjay Corporation, Linden, N. J.

Tricosene-11, a compound with a centrally located double bond, similar to methyl oleate, yielded a mixture of acid and acid chloride. 2-Methyl-2-butene, 2-pentene, and 4-methyl-2-pentene also yielded such mixtures, the relative compositions of which are summarized in Table III. The yields of phosphorus derivatives obtained from simpler, less branched olefins, such as cyclohexene or *alpha*-olefins, were lower than with other olefins.

In all cases, the degree of residual unsaturation and the amount of chlorine in the molecule indicated two things: (1) Amount of olefin reacted (loss of unsaturation); (2) extent of hydrolysis of the phosphorus acid chloride during work-up. By running chlorine analyses and saponification equivalents, the amount of acid and acid chloride structure present could be estimated.

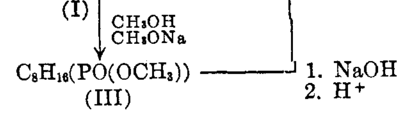
To gain a better understanding of the structure of the phosphorylated products obtained from the aluminum chloride catalyzed addition of phosphorus trichloride to olefins, 2,4,4-trimethylpentene-2 was used as a model compound. A crystalline material with a sharp reproducible melting point was obtained in high yield (90%) under the standard reaction conditions. The material had a neutral equivalent and an iodine value of zero, and elemental analyses gave the following empirical formula: $C_8H_{16}POCl$(I). The presence of only eight carbons indicated that the methanol used for the decomposition of the complex did not participate in this particular reaction. This was confirmed by repeating the general procedure, but omitting step (3), that is reaction with methanol; the identical product was obtained.

The chlorine is readily replaced by a hydroxyl group by either refluxing it with water or base or by heating to 100° in an open beaker. The resultant product is a monobasic phosphorus containing acid (II). Reaction with thionyl chloride changed the acid (II) back to the acid chloride (I), as demonstrated by a mixed melting point determination.

Also isolated during the crystallization of the original acid chloride (I) was a small amount (3–5%) of a second crystalline material, which was neutral and free of chlorine. Complete elemental analyses and hydrolysis experiments showed it to be the methyl ester, 

$$\mathrm{R}\left\langle \begin{array}{l}\mathrm{P}\rightarrow\mathrm{O}\\ \quad\mathrm{OCH_3}\end{array}\right)$$

The methyl ester was also prepared by treating the acid chloride with sodium methoxide in methanol. The transformations are summarized:

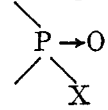
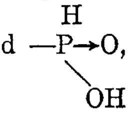
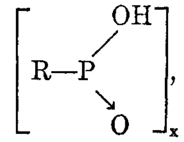


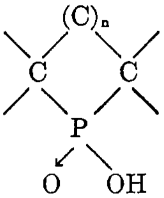
$$\begin{tikzcd}
C_8H_{16} \arrow[r, "PCl_3-AlCl_3", "CH_3OH,\ H_2O"'] & C_8H_{16}(POCl) \arrow[r, shift left, "H_2O"] \arrow[d, "CH_3OH\ CH_3ONa", "(I)"'] & C_8H_{16}(PO(OH))\ (II) \arrow[l, shift left, "SOCl_2"] \\
 & C_8H_{16}(PO(OCH_3))\ (III) \arrow[ru, "1.\ NaOH,\ 2.\ H^+"'] &
\end{tikzcd}$$

The exact attachment of the phosphorus group to the carbon skeleton has been studied for this model

TABLE III

Olefin	% >RPOCl	% >RPOOH	P Calcd.	P Found	Cl, Found	Sap. Equiv., Found
2-Methyl-2-butene	26.6	48.4	16.73	17.4	6.78	150
2-Pentene	29.1	37.8	15.60	17.4	6.22	140
4-Methyl-2-pentene	28.5	63.5	18.75	17.8	6.06	129.5

compound. All the data reported in this paper indicated presence of a  grouping, where X is either Cl, OH, or $—OCH_3$. To fulfil the normal valence requirements of phosphorus, several possibilities exist: The compounds could be derivatives of phosphorous acid , or polymers of the type , or cyclic derivatives where two of the phosphorus valences are satisfied by a carbon skeleton derived from the starting olefin, *e.g.*



The first possibility has been ruled out, since the compounds do not give reactions typical of a —P—H bond. Molecular weight determinations do not support the polymer concept. The cyclic structure is most favored for these compounds; thus far it has been possible to support such a structure in only one case, 2,4,4-trimethyl-2-pentene, where crystalline derivatives were isolated. These were proven to be 4-membered phosphorus containing ring compounds.[11] It has not yet been possible to extend proof of such a cyclic structure to all the other olefins cited, though all form a carbon-phosphorus bond and a P→O group. There are preliminary indications that only olefins with hindered double bonds give rise to cyclic structures. The purpose of the present paper is to indicate the general scope of this new reaction for forming carbon-phosphorus bonds with olefins.

EXPERIMENTAL

Three typical examples of the general phosphorylation procedure are shown: 1. *Methyl oleate.* The reaction was run in a round bottom flask with a stirrer, thermometer, dropping funnel, and a condenser with a drying tube. To the reaction flask was added 125 ml. of methylene chloride, 42.8 g. (0.316 mole) of phosphorus trichloride, and 41.6 g. (0.316 mole) of aluminum chloride. The mixture was cooled to 0° and 46.2 g. (0.158 mole) of methyl oleate in 62.5 ml. of methylene chloride was added over a 45-min. period. During the addition, the temperature was kept at 0°. After the addition was completed, stirring was continued at room temperature for 1.5 hr. Anhydrous methanol (250 ml.) was then added over a half-hour period, allowing the temperature to rise to 40°. The reaction mixture was then stirred for 2 hr. at room temperature. Excess water was carefully added with stirring. The organic layer was then separated, washed with water until neutral, and dried over anhydrous sodium sulfate. The methylene chloride was taken off *in vacuo* and 61 g. of a light yellow oil was obtained.

Anal. Calcd. for $C_{19}H_{36}PO_3Cl$: P, 8.2; Cl, 9.4; Sap. Equiv., 189. Found: P, 8.3; Cl, 3.6; sap. equiv., 210. The low values for Cl indicated that extensive hydrolysis of the acid chloride to the acid form had occurred.

2. *2,4,4-Trimethylpentene-2.* Using a similar procedure as above, 33.7 g. (0.3 mole) of 2,4,4-trimethylpentene-2 was dissolved in 100 ml. of methylene chloride and treated with 53.3 g. (0.4 mole) of aluminum chloride and 55 g. (0.4 mole) of phosphorus trichloride in 200 ml. of methylene chloride at 0°. After 30 min., 320 ml. of methanol was added, allowing the reaction mixture to warm up to 20°. Cold water was added and the organic layer separated. After drying over anhydrous sodium sulfate, the methylene chloride was taken off slowly *in vacuo.* As the volume of solvent decreased, a neutral crystalline material separated out which was isolated. The yield was 51.1 g. (88% based on $C_8H_{16}POCl$). This material was recrystallized from petroleum ether (b.p. 60–68°) and gave a material, m.p. 74–75°.

Anal. Calcd. for $C_8H_{16}POCl$: C, 49.36; H, 8.29; P, 15.92; Cl, 18.22. Found: C, 49.80; H, 8.45; P, 15.90; Cl, 18.20.

Hydrolysis yielded a monobasic acid (II), m.p. 75–76°. Calcd. for $C_8H_{16}PO_2H$: C, 54.53; H, 9.73; P, 17.58; neut. equiv., 176.2. Found: C, 54.89; H, 9.72; P, 16.07; neut. equiv., 176.2.

3. *Commercial nonene* (propylene trimer). Using the same procedure as above, commercial nonene (Enjay Co., N. J.) was treated using the following quantities: 25.2 g. (0.2 mole) of nonene dissolved in 80 ml. of methylene chloride, 53.3 g. (0.4 mole) of aluminum chloride, 55 g. (0.4 mole) of phosphorus trichloride in 160 ml. of methylene chloride, and 320 ml. of methanol. During the addition of methanol, the reaction mixture was kept between 15–20° by cooling and not allowed to warm up as in the case of the methyl oleate reaction. The yield was 39.8 g. [95% based on $C_9H_{18}(POCl)$].

Anal. Calcd. for $C_9H_{18}POCl$: P, 14.9; Cl, 17.0. Found: P, 13.9; Cl, 15.3. Hydrolysis yielded a chlorine free acid. Calcd. for $C_9H_{18}PO_2H$: neut. equiv., 190.6. Found: neut. equiv., 210.

Other olefins yielded the following phosphorylated products:

Phosphorylated triisobutylene.[12] Viscous oil. *Anal.* Calcd. for $C_{12}H_{24}POCl$: P, 13.7; Cl, 15.7; iodine value, 0.0. Found: P, 11.1; Cl, 11.7; iodine value, 29.7. This is a crude

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[11] J. J. McBride, Jr., Eric Jungermann, J. V. Killheffer, and R. J. Clutter, "A Novel Synthesis of a 4-Membered Phosphorus-Containing Ring Compound," presented before the Organic Division at the 140th National Meeting of the ACS, Chicago, Ill., September 1961.

[12] Mixture of 2,2,4,6,6-pentamethyl-3-heptene and 2-neopentyl-4,4-dimethyl 1-pentene.

reaction product. The high iodine value and low P and Cl results indicated that unreacted olefin is present in the product.

Phosphorylated Pentadecene.[13] Viscous oil. *Anal.* Calcd. for $C_{15}H_{30}POCl$: P, 11.1; Cl, 12.8; iodine value, 0.0. Found: P, 9.07; Cl, 9.15; iodine value, 30.4. This is a crude reaction product. The high iodine value, and low P and Cl results indicated that unreacted olefin is present in the product.

Tricosene-11: Equimolar quantities of olefin, PCl_3, and $AlCl_3$ were used in this reaction. Obtained a viscous oil. Calcd. for $C_{23}H_{46}POCl$: P, 7.65; Cl, 8.77; iodine value, 0.0. Found: P, 7.8; Cl, 4.9; iodine value, 9.0.

The following olefins were obtained as mixtures of free acid and acid chloride. The relative composition is based on % Cl and saponification equivalent determinations. Results are shown in Table III.

With simpler olefins, such as *alpha* olefins and cyclohexene, low yields were obtained as indicated by high iodine values and low P and Cl analyses. Removal of the starting olefins was difficult, since some polymerization had occurred. Cyclohexene is cited as a typical example. The product obtained is a viscous oil with the following analysis: Calcd. for $C_6H_{12}POCl$: P, 18.86; Cl, 21.55; iodine value, 0.0. Found: P, 13.7; Cl, 8.9; iodine value, 41.

General procedure for preparing esters from phosphorylated olefins. The crude phosphorylation product was added dropwise with stirring to a solution containing a 10–20% excess of sodium alkoxide in the corresponding alcohol at 50–60° over a period of about 45 min. The mixture was stirred for 15–30 min. longer. After cooling to room temperature the product was taken up in ether, the phases separated and the ether solution washed with water until the washings were neutral. After drying, the ether and excess alcohol were removed *in vacuo*. Yields of crude ester based on starting olefin varied from 55 to 70% depending on the olefin. If the intermediate acid chloride was purified before use, as can readily be done with the 2,4,4-trimethylpentene phosphorus-acid chloride derivative, yields were nearly quantitative.

Methyl ester of phosphorylated 2,4,4-trimethylpentene-2. Colorless liquid or white solid, m.p. 35–36; b.p. 78°/0.7 mm.

Anal. Calcd. for $C_8H_{16}PO_2CH_3$: sap. equiv., 190.2; P, 16.3; mol. wt., 190.2. Found: sap. equiv., 190.6; P, 17.3; mol. wt. (ebullioscopic in acetone), 201.

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[13] Commercial grade propylene pentamer with 3–4% terminal olefin.

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, UNIVERSITY OF NEW HAMPSHIRE]

1,2-Diacyloxyditins: A New Class of Organotin Compound[1]

ALBERT K. SAWYER AND HENRY G. KUIVILA

Received May 2, 1961

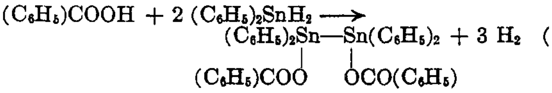
In the reactions of diphenyltin dihydride with carboxylic acids, 1,1,2,2-tetraphenyl-1,2-diacyloxyditins are formed. With di-*n*-butyltin dihydride and carboxylic acids either this type of ditin or simple di-*n*-butyldiacyloxytin is formed, depending on the acid and the acid/hydride ratio. Quantitative yields of 1,2-diacyloxyditins are also obtained in reactions of di-*n*-butyltin dihydride with di-*n*-butyldiacyloxytins in equimolar ratio. Reactions between bromine and 1,2-diacyloxyditins may be used for quantitative determination of the latter and lead to the formation of disubstituted acyloxybromotins.

In an attempt to prepare diphenyltin dibenzoate by reaction 1, (analogous to reaction 2 observed in earlier work[2]), less than the expected quantity of

$$2\,(C_6H_5)COOH + (C_6H_5)_2SnH_2 \longrightarrow (C_6H_5)_2Sn(OCO(C_6H_5))_2 + 2\,H_2 \quad (1)$$

$$2\,HCl + (C_6H_5)_2SnH_2 \longrightarrow (C_6H_5)_2SnCl_2 + 2\,H_2 \quad (2)$$

hydrogen, along with a crystalline product with analysis corresponding to 1,1,2,2-tetraphenyl-1,2-dibenzoyloxyditin was obtained. The structure of the solid product was assigned on the basis of the elemental analysis, the stoichiometry of Equation 3, and the reactions discussed below.



$$2\,(C_6H_5)COOH + 2\,(C_6H_5)_2SnH_2 \longrightarrow \begin{array}{cc} (C_6H_5)_2Sn & \!\!\!—Sn(C_6H_5)_2 \\ | & | \\ (C_6H_5)COO & OCO(C_6H_5) \end{array} + 3\,H_2 \quad (3)$$

In studying the scope of this reaction, carboxylic acids of increasing strength up to trifluoroacetic acid were used with diphenyltin dihydride. The results are shown in Table I. Although in most cases the carboxylic acid was used in excess of that required by reaction 3, the only product isolated was the ditin, and whenever the hydride was used in excess the yield of pure ditin was higher. Although other cases gave nearly the expected amount of hydrogen by reaction 3, trifluoroacetic and trichloroacetic acids gave only about one third of this amount even though all hydride was consumed. It is probable that the trihalomethyl groups were partially reduced by the hydride, as it has been shown that organotin hydrides reduce aliphatic and aromatic halides, the former readily and the latter with more difficulty.[3]

In extending this reaction to di-*n*-butyltin dihydride it became difficult to get crystalline products, and in the case of succinic acid a product

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[2] H. G. Kuivila, A. K. Sawyer, and A. G. Armour, *J. Org. Chem.*, **26**, 1426 (1961).

[3] (a) J. G. Noltes and G. J. M. van der Kerk, *Functionally Substituted Organotin Compounds*, Tin Research Institute, Greenford, Middlesex, England, 1958; G. J. M. van der Kerk and J. G. A. Luijten, *J. Applied Chem.*, **7**, 356 (1957). (b) C. A. Rothman and E. I. Becker, *J. Org. Chem.*, **24**, 294 (1959).