449. *Tropylium Complex Halide Salts of Boron, Aluminium, Iron, Titanium, and Tin.*

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Interaction of cycloheptatriene and t-butyl chloride in the presence of BCl_3, Al_2Cl_6, Fe_2Cl_6, and $SnCl_4$, respectively, gives the tropylium salts $C_7H_7^+BCl_4^-$, $C_7H_7^+AlCl_4^-$, $C_7H_7^+FeCl_4^-$, and $(C_7H_7)_2^{2+}SnCl_6^{2-}$. The use of titanium tetrachloride gives a product believed to be $C_7H_7^+TiCl_5^-$. Silicon tetrachloride shows very slight catalytic activity. Reaction between cycloheptatriene and t-butyl chloride does not occur in the absence of catalysts. Salts of the above type are also formed by metathesis, *e.g.*, between $C_7H_7^+Br^-$ and $(NH_4)_2^{2+}SnCl_6^{2-}$, or by direct addition, *e.g.*, of $C_7H_7^+Cl^-$ to $SnCl_4$.

ABSTRACTION of hydride from the methylene group in cycloheptatriene, C_7H_8, gives the tropylium ion, $C_7H_7^+$. Dauben, Gadecki, Harmon, and Pearson [1] accomplished this in the reaction of cycloheptatriene with triphenylmethyl perchlorate and fluoroborate in acetonitrile to give the corresponding tropylium salts. Triphenylmethyl halides were reported not to act as satisfactory hydride-acceptors in acetonitrile, but gave high yields of tropylium halides in liquid sulphur dioxide. Halide-hydride exchange, recently reviewed,[2] is known to be often catalysed by acids. For example, Bartlett, Condon, and Schneider [3] showed that aluminium bromide catalysed such exchange between t-butyl chloride and isopentane or isobutane. Volpin and his co-workers [4] have reported extensive studies on the interaction between cycloheptatriene and various acids and inorganic halides to give tropylium salts, some in high yields; but few preparative details and analyses of products have so far been recorded.

The present paper deals with halide-hydride exchange between cycloheptatriene and t-butyl chloride in the presence of various inorganic halides. No uncatalysed reaction between cycloheptatriene and t-butyl chloride could be detected; but in the presence of those halides which can function as Lewis acids, tropylium salts were readily formed. Thus stannic chloride (cf. Bryce-Smith and Perkins [5]), stannic bromide, aluminium chloride, boron trichloride, and ferric chloride, respectively, gave the following salts: $(C_7H_7)_2^{2+}SnCl_6^{2-}$, $(C_7H_7)_2^{2+}SnBr_6^{2-}$ (from t-butyl bromide), $C_7H_7^+AlCl_4^-$, $C_7H_7^+BCl_4^-$, and $C_7H_7^+FeCl_4^-$. The stannichloride was a colourless solid, essentially insoluble in water,

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[1] Dauben, Gadecki, Harmon, and Pearson, *J. Amer. Chem. Soc.*, 1957, **79**, 4557.
[2] Deno, Peterson, and Saines, *Chem. Rev.*, 1960, **60**, 7.
[3] Bartlett, Condon, and Schneider, *J. Amer. Chem. Soc.*, 1944, **66**, 1531.
[4] (a) Kursanov and Volpin, *Doklady Akad. Nauk S.S.S.R.*, 1957, **113**, 339; (b) Kursanov, Volpin, and Parnes, *Khim. Nauk i Prom.*, 1958, **3**, 159; (c) Volpin, *Uspekhi Khim.*, 1960, **29**, 298.
[5] Bryce-Smith and Perkins, *Chem. and Ind.*, 1959, 1022.

ethanol, ether, acetone, and hydrocarbons. It was not hygroscopic and showed no tendency to react with oxygen-containing media. The stannibromide was yellow, but otherwise similar to the stannichloride (cf. tropylium bromide and chloride[6]). The colourless deliquescent tetrachloroaluminate resembled the corresponding alkali-metal salts[7] in its failure to form complexes of higher co-ordination number under the conditions employed. Its freshly-prepared aqueous solution was pink, but the colour slowly faded; the pink solution in ethanol did not fade. It was insoluble in ether and hydrocarbons, and did not form an etherate. The colourless deliquescent tetrachloroborate was soluble in ethanol, but insoluble in ether and hydrocarbons, and did not form an etherate. The yellow tetrachloroferrate was not deliquescent, but was freely soluble in water and acetone, sparingly soluble in boiling ethanol, and insoluble in ether and hydrocarbons. All of these salts were soluble in dilute hydrochloric acid, and the solutions showed the ultraviolet spectrum of the tropylium ion.[6]

Titanium tetrachloride, t-butyl chloride, and cycloheptatriene readily gave a yellow solid, which turned colourless in air, gave colourless non-reducing solutions in water or ethanol, and when treated with ether gave a colourless ether-insoluble product, probably an etherate. The yellow compound was insoluble in hydrocarbons. Its aqueous solution was shown to contain tropylium ions by the ultraviolet spectrum,[6] the reaction with sodium tetraphenylboron to give tropylium tetraphenylboron,[8] and the formation of di(cycloheptatrienyl) ether by the action of sodium hydrogen carbonate. Elemental analysis proved extremely difficult, but structures such as the expected $(C_7H_7)_2^{2+}TiCl_6^{2-}$, or $C_7H_7TiCl_3$ were ruled out. The compound is probably $C_7H_7^+TiCl_5^-$. The pentachlorotitanate ion is novel, and is isoelectronic with the as yet unknown vanadium pentachloride. Vanadium pentafluoride is well known, as are the pentachlorides of niobium and tantalum. A search of the literature has revealed only one previous example of quinqueco-ordinate titanium, *viz.*, the $TiCl_5^{2-}$ ion, which Erlich *et al.*[9] have shown to exist in the system KCl–$TiCl_3$.

A mixture of silicon tetrachloride, t-butyl chloride, and cycloheptatriene in hexane very slowly formed tropylium ions, although no definite complex was isolated. Tropylium ions were not found when phosphorus trichloride was used in place of silicon tetrachloride. It seems possible that catalysis of the formation of tropylium ion from cycloheptatriene in the presence of t-butyl chloride may provide a very sensitive test for a Lewis acid.

All the above complex halides were also prepared by direct addition of a tropylium halide to the appropriate inorganic halide, *e.g.*, $2C_7H_7^+Cl^- + SnCl_4 \longrightarrow (C_7H_7)_2^{2+}SnCl_6^{2-}$. In addition, tropylium stannichloride was prepared by double decomposition of tropylium bromide and ammonium stannichloride in alcohol.

Since the activity of the foregoing inorganic halides in promoting the formation of tropylium salts from cycloheptatriene and t-butyl chloride appears related qualitatively to their acceptor properties, *i.e.*, to their activity as Lewis acids, it seems probable that the overall reaction may be an acid-catalysed halide-hydride exchange. In the case (say) of boron trichloride, the following mechanism appears plausible (cf. ref. 4):

$$C_7H_8 + BCl_3 \rightleftharpoons C_7H_7^+BHCl_3^- \quad . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \quad (1)$$

$$C_7H_7^+BHCl_3^- + Bu^tCl \longrightarrow C_7H_7^+BCl_4^- + Bu^tH \quad . \; . \; . \; . \; . \; . \; . \; . \quad (2)$$

The question whether tropylium halides were formed as intermediates cannot be definitely decided on the evidence available. Were they so formed, which we think improbable, it is established that reaction with free inorganic halide would give the complex salts actually isolated. In the absence of t-butyl chloride, stannic chloride and cycloheptatriene gave a very low yield of tropylium stannichloride. This suggests that stannic

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[6] Doering and Knox, *J. Amer. Chem. Soc.*, 1954, **76**, 3203.

[7] Malquori, *Atti Accad. naz. Lincei, Rend. Classe Sci. fis. mat. nat.*, 1927, **5**, 510; 1928, **7**, 745.

[8] Volpin, Zhdanov, and Kursanov, *Doklady Akad. Nauk S.S.S.R.*, 1957, **112**, 264.

[9] Erlich, Kaupa, and Blankerstein, *Z. anorg. Chem.*, 1959, **299**, 213.

chloride itself, or some chlorine-containing by-product, can act as chloride donor as in equation (2) above. Allyl chloride was much less effective than t-butyl chloride as a chloride-donor.

Hydrolysis of Tropylium Stannichloride.—Cycloheptatrienol apparently unknown pure, is stated to form di(cycloheptatrienyl) ether at high pH.[6] In accordance with this, tropylium stannichloride was readily decomposed by cold aqueous sodium hydroxide to give moderate yields of the ether. Higher yields were obtained by the use of cold aqueous sodium hydrogen carbonate, but the reason for this is not clear. Bickel [10] found, and we have confirmed, that the ether is stable to aqueous sodium hydroxide, and that this reagent serves to remove the cycloheptatrienone which is liable to be formed when acidic conditions exist during the hydrolysis of tropylium salts.[11,12] We found that the ether formed by all our procedures of alkaline hydrolysis usually contained a few per cent. of the ketone, as recognised by the absorption peak at 6·3 μ in the infrared spectrum. This was eventually traced to the use of acid-washed glassware in the final distillation stage. The use of alkali-washed glassware gave the ether completely free from cycloheptatrienone.

[*Added in proof.*] We have now found that Kursanov and Volpin's supposed preparation of tropylium chloride [4a] in fact gives a complex tropylium salt which contains phosphorus. This is similar to tropylium chloride in several physical and chemical properties, and is under investigation. A few of the results in this paper are affected, but not to an important extent since the compositions of the products have been determined without reference to the starting materials.

EXPERIMENTAL

Materials.—t-Butyl chloride was freshly fractionated before use and had b. p. 50—51°. t-Butyl bromide was prepared and purified as described by Bryce-Smith and Howlett.[13] Silicon tetrachloride, b. p. 57—58°; stannic chloride, b. p. 112—114°; and titanium tetrachloride, b. p. 134—137°, were refractionated before use. The other inorganic halides were commercial materials of good quality.

Cycloheptatriene (International Shell Petroleum Company) was about 93% pure after redistillation. Yields have not been corrected for this. Traces of a yellow compound invariably codistilled, but could be removed by treatment with sodium methoxide or ammonia. The colour arose from a small peak at *ca.* 357 mμ which tailed into the visible. This, coupled with the sensitivity to bases, suggests that the impurity was a methylfulvene: cf. the reported [14] $\lambda_{max.} = 355$ mμ. All solvents were dried before use, and anhydrous conditions were maintained during experiments as far as possible. All compounds formulated as tropylium salts showed the ultraviolet spectrum of the tropylium ion in 2N-hydrochloric acid.[6]

Preparation of Tropylium Stannichloride.—(a) *From cycloheptatriene, t-butyl chloride, and stannic chloride.* Experimental details and analytical results have been given.[5] The ultraviolet absorption data recorded therein for the 0·005% solution in 2N-hydrochloric acid were incorrectly calculated, and should be replaced by the following values: $\lambda_{min.}$ 258—259 mμ (log $\varepsilon_{min.}$ 3·34); $\lambda_{max.}$ 276—277 mμ (log $\varepsilon_{max.}$ 3·61). The m. p. was 252° (decomp.), and the yield 33% of theoretical.

(b) *From cycloheptatriene, allyl chloride, and stannic chloride.* The use of an equivalent proportion of allyl chloride in place of t-butyl chloride in (a) gave a 5% yield of tropylium stannichloride, m. p. 248° (decomp.) (Found: Sn, 23·2; Cl, 40·8. Calc. for $C_{14}H_{14}Cl_6Sn$: Sn, 23·1; Cl, 41·45%).

(c) *From cycloheptatriene and stannic chloride.* Stannic chloride (19·5 g., 0·075 mole) was added with stirring during 20 min. to a refluxing mixture of cycloheptatriene (13·8 g., 0·15 mole) and hexane (75 ml.). The resulting dark yellow solution was heated under reflux for a further 4½ hr. Filtration, followed by washing of the solid with dry ether (Soxhlet), gave tropylium stannichloride (0·97 g., 2·5% of theoretical), m. p. 251° (decomp.) [Found: C, 33·6; H, 2·95;

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[10] Bickel, private communication.

[11] Ter Borg, van Helden, Bickel, Renold, and Dreiding, *Helv. Chim. Acta*, 1960, **43**, 457.

[12] Ikemi, Nozoe, and Sugiyama, *Chem. and Ind.*, 1960, 932.

[13] Bryce-Smith and Howlett, *J.*, 1951, 1141.

[14] Angus, Blair, and Bryce-Smith, *J.*, 1960, 2003.

Cl, 40·6; Sn, 22·9. Calc. for $C_{14}H_{14}Cl_6Sn$: C, 32·7; H, 2·75; Cl, 41·45; Sn, 23·1%. This analysis rules out any possibility that the product was $(C_7H_7)_2{}^{2+}SnCl_4H_2{}^{2-}$, which would require C, 37·8; H, 3·6; Cl, 31·9; Sn, 26·7%; but the presence of traces of such a salt would largely account for the mediocre analytical data].

(d) *From tropylium bromide and ammonium stannichloride.* A solution of tropylium bromide [15] (3·0 g.) in ethanol (35 ml.) was added dropwise to a hot solution of ammonium stannichloride (3·22 g.) in ethanol (300 ml.). The off-white precipitate was purified as in method (c) to give tropylium stannichloride (2·18 g., 48·5%), m. p. 253° (decomp.) (Found: C, 32·4; H, 2·7; Cl, 40·9; Sn, 23·1%).

(e) *From tropylium chloride and stannic chloride.* A solution of tropylium chloride [4a] (1·52 g., 0·012 mole) in ethanol (45 ml.) was added with stirring at room temperature to a solution of stannic chloride (1·56 g., 0·006 mole) in ethanol (45 ml.). The white precipitate which immediately formed was filtered off and washed successively with ethanol and ether (yield 2·27 g., 74%), m. p. 254° (decomp.) (Found: C, 32·9; H, 2·8; Cl, 40·6; Sn, 23·0%).

Preparation of Tropylium Stannibromide.—(a) *From cycloheptatriene, t-butyl bromide, and stannic bromide.* The procedure closely followed (a) above for the stannichloride, except that equivalent proportions of bromides were used in place of chlorides. The *product*, m. p. 249° (decomp.), a yellow solid, insoluble in water and common organic solvents, was obtained in 12% yield (Found: C, 21·4; H, 1·7; Br, 60·7; Sn, 15·2. Calc. for $C_{14}H_{14}Br_6Sn$: C, 21·6; H, 1·8; Br, 61·4; Sn, 15·2%).

(b) *From tropylium bromide and stannic bromide.* A solution of stannic bromide (1·1 g., 0·0025 mole) in ethanol (30 ml.) was added to a solution of tropylium bromide (0·86 g., 0·005 mole) in ethanol (30 ml.). The yellow precipitate was filtered off and washed successively with ethanol and ether. It had m. p. 253° (decomp.) (yield 1·68 g., 86%) (Found: C, 21·4; H, 2·5; Br, 61·8; Sn, 15·1%.)

The compounds $(C_7H_7)_2{}^{2+}SnBr_2Cl_4{}^{2-}$ and $(C_7H_7)_2{}^{2+}SnBr_4Cl_2{}^{2-}$ were similarly prepared by direct addition of the corresponding halides in ethanol; they had m. p.s 242° and 246°, respectively. The compositions were confirmed by elemental analyses. The physical properties were closely similar to those of the other stannihalides.

Preparation of Tropylium Tetrachloroborate.—(a) *From cycloheptatriene, t-butyl chloride, and boron trichloride.* A mixture of cycloheptatriene (18·4 g.), t-butyl chloride (18·6 g.), boron trichloride (23·5 g.), and hexane (50 ml.) was stirred at room temperature for 6 hr. (cold-finger condenser at −70°). The white precipitate was extracted with ether (Soxhlet) and dried at room temperature *in vacuo*. The *product* (2·8 g., 6%), m. p. 180—190° (decomp.: rather indefinite), was deliquescent, and soluble in water, acetone, and ethanol, but insoluble in ether and hydrocarbons (Found: C, 34·8; H, 3·3; Cl, 57·9. $C_7H_7Cl_4B$ requires C, 34·5; H, 2·9; Cl, 58·2%).

(b) *From tropylium chloride and boron trichloride.* A mixture of tropylium chloride (0·75 g.) and boron trichloride (0·8 g.) was kept at room temperature for 4 days in a sealed tube under nitrogen. Ether extraction (Soxhlet) of the product gave tropylium tetrachloroborate (1·19 g., 82%) as a colourless solid, m. p. *ca.* 180° (Found: C, 34·8; H, 3·3%).

Preparation of Tropylium Tetrachloroaluminate.—(a) *From cycloheptatriene, t-butyl chloride, and aluminium chloride.* A mixture of cycloheptatriene (9·2 g., 0·1 mole), t-butyl chloride (9·3 g., 0·1 mole), and aluminium chloride (13·4 g., 0·05 mole) was heated under reflux with stirring under argon for 1 hr., and then cooled. Pentane (50 ml.) was added. The buff-coloured paste was filtered off, washed with pentane, and while still damp with pentane was quickly transferred to a Soxhlet apparatus. Extraction with ether for 14 hr. left a colourless solid. Last traces of ether were removed under reduced pressure to give the highly deliquescent *tropylium tetrachloroaluminate* (5·64 g., 22%), m. p. 208° (decomp.) (Found: C, 31·5; H, 3·7; Al, 10·1; Cl, 52·4. Calc. for $C_7H_7AlCl_4$: C, 32·3; H ,2·7; Al, 10·4; Cl, 54·6%. $C_7H_7AlCl_4$ containing 3% of H_2O requires C, 31·4; H, 3·0; Al, 10·1; Cl, 52·9%). The compound was freely soluble in water and ethanol, and insoluble in ether and hydrocarbons.

(b) *From tropylium chloride and aluminium chloride ethanolate.* A solution of aluminium chloride (1·3 g., 0·005 mole) in ethanol (30 ml.) was added with stirring at room temperature to a solution of tropylium chloride (1·26 g., 0·01 mole) in ethanol (20 ml.). The mixed solution was kept in a desiccator for 2 hr., and then ether (200 ml.) was added. By the procedure

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[15] Doering and Knox, *J. Amer. Chem. Soc.*, 1957, **79**, 352.

described for the previous experiment, tropylium tetrachloroaluminate (1·78 g., 70%) was obtained. As in the previous case, the highly deliquescent nature of this material hindered accurate analysis (Found: C, 30·3; H, 3·5; Al, 10·2; Cl, 52·7. Calc. for $C_7H_7AlCl_4$, containing 6% of H_2O: C, 30·4; H, 3·2; Al, 9·8; Cl, 51·3%).

Preparation of Tropylium Tetrachloroferrate.—(a) *From cycloheptatriene, t-butyl chloride, and ferric chloride.* Ferric chloride (16·2 g.) was added with stirring to a mixture of cycloheptatriene (18·4 g.) and t-butyl chloride (19·0 g.). Hydrogen chloride was soon evolved, and the temperature was kept below 50° by cooling. When the evolution of heat ceased, the product was stirred at 65° for 5 hr. and then cooled. Pentane (50 ml.) was added and the whole product was transferred to a 100-ml. measuring cylinder. Unchanged ferric chloride rapidly settled and the supernatant yellow suspension was decanted. This procedure was repeated twice more. The final suspension was filtered and the yellow residue washed with ethanol (3 × 30 ml.) and finally with pentane.

Tropylium tetrachloroferrate (3·0g., 5%) was obtained as a yellow solid, m. p. 227° (Found: C, 29·5; H, 2·6; Cl, 48·6; Fe, 19·0. Calc. for $C_7H_7Cl_4Fe$: C, 29·1; H, 2·5; Cl, 49·1; Fe, 19·3%). The compound was stable in moist air, freely soluble in water and acetone, sparingly soluble in ethanol, and insoluble in ether and hydrocarbons.

(b) *From tropylium chloride and ferric chloride ethanolate.* A solution of ferric chloride (1·62 g., 0·005 mole) in ethanol (20 ml.) was added with stirring to a solution of tropylium chloride (1·26 g., 0·01 mole) in ethanol (50 ml.). After 1 hr. the yellow solid was filtered off, washed with ethanol (3 × 10 ml.) and then pentane, giving tropylium tetrachloroferrate (2·43 g., 84%), m. p. 228° (Found: C, 29·4; H, 2·7; Cl, 48·9; Fe, 19·2%).

Reaction of Cycloheptatriene, t-Butyl Chloride and Silicon Tetrachloride.—A mixture of the three components (9·2 g., 9·3 g., and 17·0 g., respectively) in hexane (50 ml.) was kept at 20° under nitrogen for 4 weeks. Traces of a brown solid separated. No pure compound was isolated, but the formation of a very small proportion of tropylium ions (0·07%) was recognised from the ultraviolet spectrum of a 2N-hydrochloric acid extract, and confirmed by the colour test with sodium tetraphenylboron.[8] A control experiment in which silicon tetrachloride was omitted gave no trace of tropylium ions.

A similar experiment with silicon tetrachloride was conducted for 12 days at 63°, and gave tropylium ions (0·03%).

Tropylium Pentachlorotitanate.—Analytical results for the products obtained by both of the following procedures are mediocre, and were obtained only with difficulty. A nitrogen atmosphere was maintained throughout each experiment.

(*a*) Titanium tetrachloride (56·8 g.) was added with stirring during 40 min. to a refluxing mixture of cycloheptatriene (55·2 g.), t-butyl chloride (55·6 g.), and hexane (400 ml.). The red-brown product was stirred under reflux for 5 hr. The buff precipitate was filtered off and washed with light petroleum (500 ml.; b. p. 40—60°). Drying *in vacuo* at 60° gave the *product* (22·35 g.) as a yellow powder, m. p. *ca.* 130° (decomp.: rather indefinite) [Found: C, 19·8; H, 2·3; Cl, 56·9; Ti, 16·4 (total 95·4%). $C_7H_7Cl_5Ti$ requires C, 26·6; H, 2·2; Cl, 56·1; Ti, 15·1%]. The tropylium ion content, estimated spectroscopically, was 25·1% (Calc. for $C_7H_7{}^+TiCl_5{}^-$: 28·8%). The low values for carbon were repeatedly confirmed (Drs. Weiler and Strauss, Oxford). The cause of the discrepancy is uncertain, but the presence of titanium may possibly have caused some interference. If the deficiency of 4·6% is assumed to be due to carbon, C : H = 0·9, which is much more in accord with the spectroscopic result than is the ratio 0·73 indicated without such correction. The structure $(C_7H_7)_2{}^{2+}TiCl_6{}^{2-}$ is ruled out (Calc. for $C_{14}H_{14}Cl_6Ti$: C, 37·9; H, 3·2; Cl, 48·0; Ti, 10·9; $C_7H_7{}^+$, 41·1%). (*b*) A mixture of tropylium chloride (1·35 g.) and titanium tetrachloride (7·3 g.) was kept at room temperature for 2 days with occasional shaking. Light petroleum (200 ml.; b. p. 60—80°) was then added. The yellow product (0·72 g.) was isolated as in the preceding experiment (Found: C, 27·1; H, 3·6; Cl, 52·8%). These values suggest that the product was contaminated with tropylium chloride, which has very similar solubility properties, and from which no complete separation could be devised. Spectroscopic determination of $C_7H_7{}^+$ gave the value 30·3%.

Di(cycloheptatrienyl) Ether.—Tropylium stannichloride (25·67 g.) was added steadily during 3 hr. to a well-stirred mixture of sodium hydrogen carbonate (42 g.), water (200 ml.), and pentane (100 ml.) at room temperature, and stirring was continued for a further 3 hr. Fractional distillation of the separated and dried ($MgSO_4$) pentane solution in alkali-washed apparatus gave di(cycloheptatrienyl) ether (6·56 g., 66%), b. p. 105—108°/0·7 mm. The absence of cycloheptatrienone was demonstrated by the absence of any peak at 6·3 μ. The compound was stable in the dark under nitrogen at 140° in alkali-washed glass, but in acid-washed glass under these conditions, cycloheptatrienone and cycloheptatriene were slowly formed (ultraviolet and infrared spectra). Cycloheptatrienone 2,4-dinitrophenylhydrazone had m. p. 177° (lit., 176—177°,[16] 165—166° [11]) and the correct analysis.

The use of equivalent proportions of sodium carbonate and sodium hydroxide (allowing for the formation of sodium stannate) in place of sodium hydrogen carbonate in the above preparation led to yields of 51 and 45%, respectively.

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Chemistry Department, The University, Reading, Berks. [*Received, December 28th,* 1960.]

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[16] Nozoe, Mukai, Takase, and Nagase, *Proc. Japan Acad.*, 1952, **28**, 477.