383. *The Preparation of Some Organotin*(IV) *Compounds.*

By D. L. Alleston and Alwyn G. Davies.

Twenty-five organotin(IV) compounds are described which are new or had not been adequately characterised. They include halides, carboxylates, alkoxides, and a series of 1 : 1 chelate complexes of 2,2′-bipyridyl or 1,10-phenanthroline with diorganotin dihalides.

Many simple organotin compounds are unknown, or have been obtained by inconvenient routes, or have been wrongly or inadequately described, usually in patents. This paper reports the preparation and properties of a number of organotin(IV) halides, alkoxides, and carboxylates, and of the complexes which the dihalides form with bidentate nitrogen ligands.

Dibutyltin dichloride in ethanol was treated with potassium fluoride, giving crystalline dibutyltin difluoride. In contrast to other known organotin fluorides it is soluble in some organic solvents and has a relatively low melting point.

Dibutyltin dibromide was obtained in 75% yield by heating together tetrabutyltin and tin tetrabromide; this is a more convenient route than the previous one[1] from dibutyltin oxide and hydrobromic acid. Dibutyltin di-iodide was prepared from the oxide and hydriodic acid.

This series of halides shows the usual decrease in melting point from fluoride to iodide.

Organotin carboxylates have been prepared previously by treating the corresponding halides with metal carboxylates[2a] although some failures have been reported.[2b] An improvement of this method is to prepare the salt *in situ* by adding the acid to a solution of sodium in an alcohol; the sodium carboxylate is obtained in a finely divided form and reacts readily with the equivalent amount of organotin halide. Alternatively, a solution of the organotin halide and carboxylic acid can be treated with an amine such as triethylamine. Both procedures are more convenient than the common one of treating the alkyltin oxide with a carboxylic acid, because the oxide is obtained by hydrolysis of the halide. Tributyltin octanoate and dibutyltin dioctanoate have been prepared by these improved methods, and dibutyltin dibenzoate by heating the compound $Bu_4Sn_2Cl_2O$[3] with benzoic acid.

It is known that rapid redistribution of halide, X and Y, occurs when different tin halides, SnX_4 and SnY_4, are mixed.[4] A similar rapid redistribution occurs between electronegative groups in dialkyltin(IV) compounds. When pentane solutions of dibutyltin dibromide and diacetate are mixed, dibutyltin acetate bromide is immediately precipitated in good yield; dibutyltin dichloride appears to react similarly with dibutyltin dibromide to give a mixed bromide chloride.

Dibutyltin acetate bromide smells of acetic acid; in the air, hydrolysis is substantial in 2 hr., and complete within 20 hr., giving acetic acid and the compound $Bu_4Sn_2Br_2O$. An attempt to prepare dibutyltin chloride octanoate from dibutyltin dichloride and sodium octanoate in butanol similarly gave butyl octanoate and the compound $Bu_4Sn_2Cl_2O$. These halide carboxylates thus readily undergo hydrolysis or alcoholysis, and apparently by a reaction involving acyl–oxygen rather than tin–oxygen bond fission. This catalysis of esterification by organotin compounds is claimed in patents relating to the preparation of carboxylic[5] and carbamic[2,6] esters.

About thirty-five organotin alkoxides are mentioned in the patent literature,[2a] but only three appear to have been analysed: diethoxydiphenyltin, which was prepared by Aronheim[7] in 1878 by reaction of diphenyltin dichloride with sodium ethoxide, and

---

[1] Pfeiffer, Lehnardt, Luftensteiner, Prade, Schnurmann, and Truskier, *Z. anorg. Chem.*, 1910, **68**, 102.

[2] (*a*) Ingham, Rosenberg, and Gilman, *Chem. Rev.*, 1960, **60**, 459; (*b*) Rosenberg, Debreczeni, and Weinberg, *J. Amer. Chem. Soc.*, 1959, **81**, 972.

[3] Alleston and Davies, *Chem. and Ind.*, 1961, 949.

[4] Burke and Lauterbur, *J. Amer. Chem. Soc.*, 1961, **83**, 326.

[5] B.P. 810,381.

[6] Cox and Hostettler, Abs. Amer. Chem. Soc. Meetings, 1959, **135**, 112-O.

[7] Aronheim, *Annalen*, 1878, **194**, 145.

tributylethoxytin and dibutyldiethoxytin which were recently obtained by Seyferth and his co-workers [8] by alcoholysis of the corresponding butylperfluorovinyltins. We have prepared tributylmethoxytin, methoxytriphenyltin, and dibutyldimethoxytin by reaction between the organotin halide and sodium alkoxide in the corresponding alcohol. The tin alkoxides are very reactive towards protic reagents and give an alcohol, which can easily be removed, as the only by-product. These alkoxides are thus convenient starting materials for preparation of further organotin compounds such as peroxides.[9]

Many complexes of organotin halides with monoamines are known,[2] but these are often rather unstable and are readily hydrolysed. We have investigated the complexes formed between dialkyltin dihalides and bidentate ligands such as 2,2′-bipyridyl and 1,10-phenanthroline, in the hope that they would be more stable. At the time that this work was carried out, no such complex had been reported, but Blake, Coates, and Tate [10] have since described the preparation of dibutyl-(2,2′-bipyridyl)tin dichloride.

When ethereal solutions of the dialkyltin dihalide and of 2,2′-bipyridyl or 1,10-phenanthroline are mixed, the complexes are precipitated in 85—98% yield.[11] They are stable and have sharp melting points (see Table), and they are convenient derivatives

2,2′-Bipyridyl and 1,10-phenanthroline complexes.

No.	R	X	Recryst. from	Yield (%)	Appearance	M. p.
				R_2SnX_2,bipy		
1	Et	Cl	C_6H_6	81	Micaceous needles	200—201°
2	Bu	Cl	C_6H_6	—	Silky needles	180 [g]
3	Bu	Br	C_6H_6	82	Small plates	176—177
4	Bu	I	EtOH	80	Pale yellow needles	163
5	Bu	CNS	BuOH	67	Lustrous needles	152·5—153
6	Oc [d]	Cl	EtOH	59	Small crystals	133—133·5
7 [a]	Ph	Cl	MeO·C_2H_4·OH [e]	72	Feathery crystals	243—245
				R_2SnX_2,phenan		
8	Et	Cl	EtOH	76	Small diamonds	235—236
9	Bu	Cl	EtOH	—	Lustrous prisms	200
10 [b]	Bu	Br	EtOH	80	Small crystals	199—200
11	Bu	I	EtOH	77	Pale yellow crystals	234—236
12	Oc [d]	Cl	EtOH	—	Lustrous needles	101 [h]
13 [c]	Ph	Cl	MeO·C_2H_4OH [e]	51	Lustrous needles	235 (decomp.)

	Found (%)					Required (%)			
No.	C	H	N	X	Formula	C	H	N	X
1	41·8	4·7	6·9	17·4	$C_{14}H_{18}Cl_2N_2Sn$	41·6	4·5	6·9	17·6
2	47·2	5·9	6·0	15·4	$C_{18}H_{26}Cl_2N_2Sn$	47·0	5·7	6·1	15·4
3	39·7	4·3	5·7	29·8	$C_{18}H_{26}Br_2N_2Sn$	39·4	4·8	5·1	29·1
4	34·2	4·6	4·0	39·2	$C_{18}H_{26}I_2N_2Sn$	33·6	4·1	4·4	39·5
5	48·3	5·1	11·3	22·9	$C_{20}H_{26}N_4S_2Sn$ [i]	47·5	5·2	11·1	23·0
6	54·0	7·5	4·9	12·4	$C_{26}H_{42}Cl_2N_2Sn$	54·6	7·4	4·9	12·4
7	53·1	3·8	5·8	15·3	$C_{22}H_{18}Cl_2N_2Sn$	52·8	3·7	5·6	14·2
8	44·6	4·3	6·4	16·5	$C_{16}H_{18}Cl_2N_2Sn$	44·9	4·2	6·5	16·6
9	49·0	5·7	6·3	14·6	$C_{20}H_{26}Cl_2N_2Sn$	49·6	5·4	5·8	14·7
10	41·7	4·7	4·9	27·8	$C_{20}H_{26}Br_2N_2Sn$	41·9	4·6	4·9	27·9
11	36·2	4·4	5·2	37·4	$C_{20}H_{26}I_2N_2Sn$	36·0	3·9	4·2	38·1
12	55·9	7·4	4·6	11·5	$C_{28}H_{42}Cl_2N_2Sn$ [j]	56·4	7·1	4·7	11·9
13	54·3	3·8	5·4	14·9	$C_{24}H_{18}Cl_2N_2Sn$	55·0	3·5	5·3	13·5

[a] I. F. G. [b] A. F. B. [c] C. G. K. [d] Oc = octyl. [e] These complexes are insoluble in common organic solvents, and chloride had to be determined by the Carius method. All the alkyl complexes are sparingly soluble in acetone, but sufficiently so for them to be titrated potentiometrically for halogen in this solvent. [f] The colour deepened when the crystals were exposed to sunlight. [g] Sublimes at 140°/0·01 mm. Blake, Coates, and Tate [10] report m. p. 179·5—180°. [h] Sinters at 73°. [i] SCN by potentiometric titration. By Carius: Found: S, 12·5. Required: S, 12·7%. [j] The infrared spectrum showed an absorption band at 3390 cm.$^{-1}$, suggesting that the compound might be hydrated. The monohydrate, $C_{28}H_{44}Cl_2N_2OSn$, requires C, 54·8; H, 7·2; Cl, 11·5; N, 4·6%.

---

[8] Seyferth, Raab, and Brändle, *J. Org. Chem.*, 1961, **26**, 2934.

[9] Alleston and Davies, unpublished work.

[10] Blake, Coates, and Tate, *J.*, 1961, 756.

[11] Alleston and Davies, *Chem. and Ind.*, 1961, 551

for isolating and characterising the organotin halides. If the complex is treated with mercuric chloride or with ferrous perchlorate, the mercuric or ferrous complex is immediately precipitated, and the dialkyltin dihalide can be recovered. This is in contrast to the report that tetrachlorobis(tripropylphosphine)tin with mercuric chloride forms a halogen-bridged complex.[12]

The bipyridyl complexes of dibutyltin dithiocyanate, butyltin trichloride, t-butyltin tribromide, and stannous chloride are also described below.

EXPERIMENTAL

Chloride, bromide, iodide, and thiocyanate attached to tin in acetone-soluble compounds were determined by potentiometric titration with silver nitrate.

Organotin Halides.—(a) *Dibutyltin di-iodide.* Dibutyltin oxide (15·0 g.) dissolved in a stirred mixture of 54% hydriodic acid (36 c.c., 1·5 mol.) and benzene (60 c.c.) during 30 min The mixture was stirred for a further hour. Next day the upper layer was washed with water and with aqueous sodium thiosulphate and dried (Na_2SO_4), yielding colourless dibutyltin di-iodide (14·9 g.), b. p. 124—134°/0·2 mm., n_D^{25} 1·6042 (Found: C, 20·0; H, 4·0; I, 51·3. Calc. for $C_8H_{18}I_2Sn$: C, 19·7; H, 3·7; I, 52·2%). Seyferth *et al.*[8] obtained the same compound, b. p. 112°/0·6 mm., n_D^{25} 1·6020, by the action of iodine on dibutyldiperfluorovinyltin.

(b) *Dibutyltin difluoride.* Potassium fluoride (2·32 g.) in water (10 c.c.) was added to a stirred solution of dibutyltin dichloride (6·08 g., 0·5 mol.) in ethanol (25 c.c.); a voluminous white solid was immediately precipitated. The mixture was diluted with water (75 c.c.) and stirred for 5 min. The solid (5·12 g., 95%) was filtered off, washed with water, dried at 120°, and recrystallised from ethanol (150 c.c.), giving *dibutyltin difluoride*, m. p. 157—160° [Found: C, 34·2; H, 6·6; Cl, 0·0; F (by hydrolysis), 13·4. $C_8H_{18}F_2Sn$ requires C, 35·5; H, 6·7; F, 14·0%]. In an attempt to prepare a bipyridyl derivative, the dibutyltin difluoride that was recovered gave a better analysis (Found: C, 35·6; H, 7·0; F, 13·9%), but had m. p. 140—144°. It was insoluble in most common organic solvents, but soluble in dimethyl sulphoxide and in dimethylformamide.

(c) *Dibutyltin dibromide.* A mixture of tetrabutyltin (34·7 g.) and tin tetrabromide (45·8 g.) was kept at 220° for 3 hr., yielding dibutyltin dibromide (59·5 g., 75%), b. p. 96—98°/0·04 mm., m. p. 20° (lit.,[1] 20°), n_D^{25} 1·5400 (Found: C, 24·8; H, 5·1; Br, 39·8. Calc. for $C_8H_{18}Br_2Sn$: C, 24·5; H, 4·6; Br, 40·7%).

Organotin Carboxylates.—(a) *Tributyltin octanoate.* Octanoic acid (7·21 g.) in absolute ethanol was added slowly to a vigorously stirred solution of sodium ethoxide (from sodium, 1·19 g.) in ethanol (100 c.c.); a thick curd of sodium octanoate was formed. Tributyltin chloride (16·3 g.) in ethanol was added slowly to the stirred suspension, giving a suspension of finely divided sodium chloride which was allowed to settle during 2 days. The supernatant solution yielded *tributyltin octanoate* (17·2 g.), b. p. 100—220°(bath)/0·005 mm., needles, m. p. 55—56°. A further crop (1·48 g.; total 88%) was obtained by extracting the sodium chloride with ethanol (Found: C, 55·4; H, 10·1. $C_{20}H_{42}O_2Sn$ requires C, 55·4; H, 9·8%). The infrared spectrum showed absorption at 1650 and 1550 $cm.^{-1}$, corresponding to CO stretching vibrations.

(b) *Dibutyltin dioctanoate.* Sodium octanoate [from octanoic acid (18·7 g.) and sodium (2·97 g.) in ethanol (200 c.c.)] was similarly treated with dibutyltin dichloride (19·7 g.) in ethanol (50 c.c.). Sodium chloride (7·11 g., 94%) was centrifuged off, and the supernatant liquid was concentrated to 40 c.c. Dry benzene (150 c.c.) was then added to precipitate the residual sodium chloride which was filtered off. The filtrate was distilled, yielding crude *dibutyltin dioctanoate* (31·2 g., 93%) as a viscous, slightly cloudy liquid. This was recrystallised from four times its volume of light petroleum at $-50°$, giving the pure product as needles which were separated off, allowed to melt, and redistilled (b. p. 177—178°/0·01 mm., n_D^{24} 1·4673) (Found: C, 55·9; H, 9·5. $C_{24}H_{48}O_4Sn$ requires C, 55·5; H, 9·3%). The infrared spectrum showed an intense band at 1600 $cm.^{-1}$.

The same product was obtained by slowly adding triethylamine (10·1 g.) in benzene (150 c.c.) to a solution of dibutyltin dichloride (15·2 g.) and octanoic acid (14·4 g.) in benzene. An exothermic reaction took place, and triethylammonium chloride (13·2 g., 96%) separated as a

---

[12] Allison and Mann, *J.*, 1949, 2915.

gelatinous precipitate. The filtrate yielded dibutyltin dioctanoate (20·2 g., 78%), b. p. 178°/0·01 mm.

(c) *Dibutyltin dibenzoate.* A mixture of the compound $Bu_4Sn_2Cl_2O$ (27·9 g.) and benzoic acid (61·0 g.) was kept at 180° for 30 min., and then distilled, yielding *dibutyltin dibenzoate* (21·1 g., 84%), b. p. *ca.* 200°(bath)/0·005 mm., m. p. 65—67° (Found: C, 55·8; H, 6·1. $C_{22}H_{28}O_4Sn$ requires C, 55·6; H, 5·9%).

Redistribution Reactions.—(a) *Dibutyltin dibromide and dibutyltin diacetate* (A. F. B. and M. H.). Solutions of dibutyltin dibromide (4 mmoles) and dibutyltin diacetate (4 mmoles), each in dry pentane (3 c.c.), were mixed, giving an immediate precipitate of dibutyltin acetate bromide, m. p. 66—66·5°, which was filtered off, washed rapidly with pentane, and dried [Found: C, 32·1; H, 5·67; Br, 21·5; OAc (by pH titration with silver nitrate), 15·9. Calc. for $C_{10}H_{21}BrOSn$: C, 32·9; H, 5·69; Br, 21·5; OAc, 15·9%]. The m. p. was not raised when the compound was recrystallised from pentane (yield, 80%). Sawyer and Kuivila [13] obtained the same compound, m. p. 67·0—68·5°, by treating 1,1,2,2-tetrabutyl-1,2-diacetoxyditin with bromine.

The course of the hydrolysis of the bromide acetate was followed by the disappearance of the strong carbonyl peak at about 1540 cm.$^{-1}$. In the air, hydrolysis was complete in 20 hr. giving the compound $Bu_4Sn_2Br_2O$, m. p. 105°.

(b) *Dibutyltin dibromide and dibutyltin dichloride* (A. F. B). The reagents were mixed in equimolar amounts and the product obtained from pentane as white crystals (71% yield), m. p. 31—32° (Found: Br, 19·5; Cl, 12·1. This corresponds to the formula $Bu_2SnBr_{0·8}Cl_{1·2}$. Calc. for $C_8H_{18}BrClSn$: Br, 22·9; Cl, 10·2%). Repeated recrystallisation of the compound from pentane had little effect on the m. p., but increased further the Cl : Br ratio. From ether it gave a bipyridyl derivative, m. p. 175—176°, in 90% yield (Found: Br, 13·1; Cl, 8·38. This corresponds approximately to the formula $Bu_2SnBr_{0·8}Cl_{1·2}$,bipy. Calc. for $C_{18}H_{26}BrClN_2Sn$: Br, 15·6; Cl, 6·92%).

Similarly, distillation of a mixture of the dichloride and the dibromide gave the bromide chloride, b. p. 98—100°/0·5 mm., m. p. 31—32° (Found: Br, 19·0; Cl, 12·3%). Johnson and Fritz [14] obtained the same bromide chloride, b. p. 104—105°/0·55 mm., m. p. 34—34·5°, by treating the compound $Bu_4Sn_2Cl_2O$ with bromine.

The chloride bromide was hydrolysed in the presence of 1 mol. of triethylamine, yielding the compound $Bu_4Sn_2Cl_2O$.

Esterification Catalysed by Organotin Compounds.—Octanoic acid (28·8 g., 0·2 mole) in dry benzene (1·1 l.) was added slowly with stirring to a warm solution of sodium butoxide (from sodium 4·60 g., 0·2 mole) in butanol (500 c.c.). A very voluminous precipitate of sodium octanoate was obtained, and the mixture was heated and stirred under reflux for 30 min. Dibutyltin dichloride (60·7 g., 0·2 mole) in dry benzene (250 c.c.) was added slowly to the stirred suspension at 35°, and the mixture was stirred for 2·5 hr. The sodium chloride (11·3 g., 97%) was centrifuged off and the solvents distilled, leaving a clear amber liquid which slowly deposited a small amount of a compound $Bu_4Sn_2Cl_2O$, m. p. 110—113°. Distillation yielded butyl octanoate (28·5 g., 71%), b. p. 55·5—63·5°/0·01 mm., n_D^{25} 1·4242, leaving a dark brown crystalline solid which was presumably $Bu_4Sn_2Cl_2O$.

Organotin Alkoxides.—In these experiments, very finely divided sodium chloride is formed which is difficult to filter off. The reactions were therefore carried out in a 250 c.c. centrifuge bottle which was fitted with a magnetic stirrer and a rubber bung carrying a dropping funnel and calcium chloride guard-tube. After the reaction, a solid rubber bung was fitted, the sodium chloride was centrifuged off at 2000 r.p.m., and the supernatant liquid was quickly decanted off in the air.

(a) *Tributylmethoxytin.* Tributyltin chloride (32·5 g.) in dry methanol (75 c.c.) was added dropwise to sodium methoxide (from sodium, 2·3 g.) in methanol (30 c.c.) at 0°, yielding sodium chloride (5·28 g., 90%), and tributylmethoxytin (26·5 g.; 85%), b. p. 97—97·5°/0·06 mm., n_D^{25} 1·4710 (Found: C, 49·4; H, 9·5. Calc. for $C_{13}H_{30}OSn$: C, 48·6; H, 9·4%). Nesmeyanov *et al.*[15] report b. p. 101—102°/2 mm., n_D^{20} 1·4745.

(b) *Methoxytriphenyltin.* By the same procedure, triphenyltin chloride (19·7 g.) in methanol (100 c.c.) and sodium methoxide (from sodium, 1·18 g.) in methanol (20 c.c.) gave sodium

---

[13] Sawyer and Kuivila, *J. Amer. Chem. Soc.*, 1960, **82**, 5958.

[14] Johnson and Fritz, *J. Org. Chem.*, 1954, **19**, 74; cf. Gibbons, Sawyer, and Ross, *ibid.*, 1961, **26**, 2304.

[15] Nesmeyanov, Lutsenko, and Ponomarev, *Doklady Akad. Nauk S.S.S.R.*, 1959, **124**, 1073.

chloride (2·85 g., 96%) and methoxytriphenyltin, m. p. (sealed tube) 63—65° (from pentane) (18·1 g., 93%) (Found: C, 56·4, 56·0; H, 4·6, 5·8. Calc. for $C_{19}H_{18}OSn$: C, 59·9; H, 4·8%. We are unable to account for this surprisingly bad analysis).

(c) *Dibutyldimethoxytin* (C. G. K.). The reaction between dibutyltin dichloride (20 g.) in methanol (22 c.c.) and sodium methoxide (from sodium, 3 g.) in methanol (29 c.c.) similarly gave sodium chloride (7·5 g., 98%) and *dibutyldimethoxytin* (12·9 g., 67%), b. p. 126—128°/0·05 mm., n_D^{25} 1·4852 (Found: C, 40·9; H, 8·1. $C_{10}H_{24}O_2Sn$ requires C, 40·7; H, 8·2%). A patent [16] reports b. p. 136—139°/1·2 mm., n_D^{20} 1·483i.

In the air the liquid was hydrolysed slowly in bulk, but very rapidly in a thin film, giving the solid oxide. In 95% ethanol, the oxide began to separate after about 3 sec.

Chelate Complexes.—(a) *From diorganotin dihalides.* A solution of the diorganotin dihalide (2 mmoles) was mixed with a solution of the ligand (2 mmoles). Ether (15—30 c.c.) was a convenient solvent in which to prepare complexes from 2,2′-bipyridyl, and ethanol (15—20 c.c.) for those from 1,10-phenanthroline monohydrate. The *complexes* separated immediately in 85—98% yield, and were recrystallised. The bipyridyl complex of dibutyltin dithiocyanate was prepared similarly. Details are given in the Table.

Dibutyl-2,2′-bipyridyltin dichloride and mercuric chloride in acetone immediately precipitated 2,2′-bipyridylmercury(II) dichloride (in 97% yield), m. p. >265° (from 2-methoxyethanol); dibutyltin dichloride (m. p. 43°) was recovered from the filtrate. Similarly ferrous and perchlorate ions in ethanol immediately precipitated the red di-(2,2′-bipyridyl)iron(II) diperchlorate.

(b) *From alkyltin trihalides.* Butyltin trichloride and bipyridyl in ether precipitated *butyl-*2,2′*-bipyridyltin trichloride* in 97% yield. It recrystallised from much butanol as a white powder, m. p. 227—228° (Found: C, 39·7; H, 4·4; Cl, 23·9; N, 6·45. $C_{14}H_{17}Cl_3N_2Sn$ requires C, 38·4; H, 3·9; Cl, 24·3; N, 6·4%). 2,2′*-Bipyridyl-t-butyltin tribromide* (M. H.) can be obtained similarly as a white powder (m. p. >300°) which is insoluble in all common organic solvents [Found: Br (Carius), 41·9. $C_{14}H_{17}Br_3N_2Sn$ requires Br, 42·0%].

(c) *From stannous chloride.* Solutions of 2,2′-bipyridyl (1·56 g., 10 mmoles) and of stannous chloride dihydrate (1·13 g., 5 mmoles) in acetone were mixed, giving an immediate bright yellow precipitate of 2,2′*-bipyridyltin*(II) *dichloride*, m. p. 214—216° (Found: C, 33·9; H, 2·5; Cl, 20·0; N, 7·9. $C_{10}H_8Cl_2N_2Sn$ requires C, 34·7; H, 2·3; Cl, 20·5; N, 8·1%). This was insoluble in all common organic solvents, but was decomposed in some on prolonged boiling, leaving a white solid.

This work was carried out during the tenure of a D.S.I.R. Fellowship by D. L. A. We are grateful to Professors E. D. Hughes, F.R.S., and Sir Christopher Ingold, F.R.S., for their interest and encouragement, and to Messrs. A. F. Bradbury, I. F. Graham, M. Hancock, and C. G. Knight who carried out some of the experimental work; in the experimental section, their individual contributions are shown by their initials.

William Ramsay and Ralph Forster Laboratories,
University College, London, W.C.1.

[*Received, November 27th,* 1961.]

---

[16] U.S.P. 2,700,675.