NOTES

772. *The Active Principles of Leguminous Fish-poison Plants. Part X. Toxicarol Isoflavone* [1]

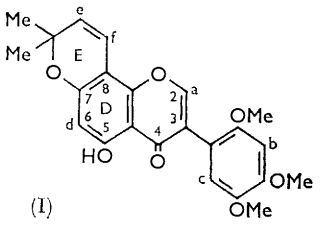
By S. H. HARPER and W. G. E. UNDERWOOD

IN 1940 one of us isolated from *Derris malaccensis* resin a phenol, isomeric with toxicarol, for which the isoflavone structure (I) was suggested.[2] With the development of spectroscopic methods of structure determination it has become possible to substantiate this structure using the small specimens remaining from the original investigation.

The infrared spectrum of the phenol showed carbonyl absorption at 1655 cm.$^{-1}$, but no phenolic hydroxyl absorption; a phenomenon observed by Flett [3] in 1-hydroxyanthraquinones, which confirms that the hydroxyl is *ortho* to the keto-group. Integrals of the signals in the nuclear magnetic resonance (n.m.r.) spectra of the acetate and the methyl ether each represent 24 protons, of which only two in each spectrum are spin coupled. The assignments are listed in the Table and were made by comparison with the spectrum of (−)-α-toxicarol.[4]

The position of the D/E ring fusion is not readily determined from the n.m.r. spectrum, but of the four possible arrangements only the two biogenetically preferred (with the oxygen at position 7, isoflavone numbering) are allowed as the single ring D proton is certainly adjacent to two oxygen functions. A negative Gibbs reaction [5] for the phenol establishes the angular fusion. The isoflavone structure is confirmed by the 2-proton in

[1] Part IX, S. H. Harper, *J.*, 1942, 595.
[2] S. H. Harper, *J.*, 1940, 1178.
[3] M. St. C. Flett, *J.*, 1948, 1441.
[4] L. Crombie and J. W. Lown, *J.*, 1962, 775.
[5] L. Crombie and R. Peace, *J.*, 1961, 5445.



(I)

Protons	Chemical shifts (δ) Methyl ether	Acetate
a	7·77	7·81
b	6·57	6·57
c	6·91	6·85
d	6·29	6·49
e	6·70 } J = 10·5	6·78 } J = 10·5 c./sec.
f	5·53 }	5·68 }
Methoxyl	3·89 (two)	3·90
,,	3·81	3·84
,,	3·71	3·75
Acetoxyl	—	2·38
gem-Dimethyl	1·48	1·49

the n.m.r. spectrum,[6] and by the ultraviolet absorption spectrum of the methyl ether which is characteristic of the unconjugated isoflavone system, $\lambda_{max.}$ 265, 294 (sh) mμ (log ε 4·63 and 4·10, respectively).

We thank Professor L. Crombie for measurement of the n.m.r. spectra.

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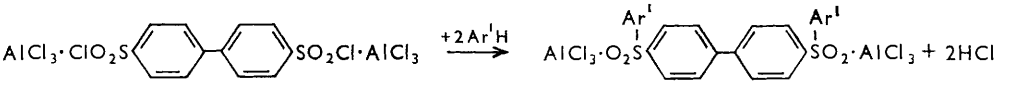
[6] Spectrum no. 696, Varian Spectra Catalog, Varian Associates, Palo Alto, 1963.

773. *Friedel–Crafts Sulphonylation with Biphenyl-4,4′-disulphonyl Chloride*

By G. Holt and K. D. Jeffreys

Although the preparation of diaryl sulphones by Friedel–Crafts sulphonylation in methylene or ethylene dichloride has been described,[1] this procedure has not been applied to disulphonyl chlorides. We now report that, when the di(aluminium chloride) complex of biphenyl-4,4′-disulphonyl chloride (I) in methylene or ethylene dichloride is added to a solution of an aromatic compound, Ar′H (2 moles), in the same solvent, there results the dioxonium complex (II) of the corresponding disulphone, from which the latter is liberated by the addition of ice and dilute hydrochloric acid.



(I) (II)

The disulphones obtained by this procedure are shown in Table 1.

Table 1

Benzene derivative	Orientation of substituents in Ar′	Yield (%)	M. p.	Ref.
C_6H_6	—	77	299—300°	2
1,4-$C_6H_4Me_2$	2,2′; 5,5′	91	243—244	
1,3,5-$C_6H_3Me_3$	2,2′; 4,4′; 6,6′	68	281—282	
$C_6H_5{\cdot}C_6H_5$	4,4′	95	361—363 (decomp.)	3

All reactions were carried out at room temperature (18—24°), over a period of 48 hr.

The solubility of 4,4′-bis(biphenyl-4″-sulphonyl)biphenyl in organic solvents was so low as to render its purification by crystallisation difficult. It was most satisfactorily purified by the addition of anisole to a solution of its oxonium complex (II; Ar′ = *p*-C_6H_4·Ph) in ethylene dichloride. This procedure precipitated the sulphone, m. p. 361—363° (decomp.), in a higher state of purity than that previously reported,[3] m. p. 345—346°.

Attempts were made to synthesise the same compounds by interaction of biphenyl and the corresponding monosulphonyl chloride (2 moles).

[1] G. Holt and B. Pagdin, *J.*, 1960, 2508.
[2] J. Huissman, U.S.P. 2,224,964/1941 (*Chem. Abs.*, 1941, **35**, 2159).
[3] H. Kuczynski, L. Kuczynski, and E. Sucharda, *Roczniki Chem.*, 1938, 625 (*Chem. Abs.*, 1940, **34**, 3246).

$$2Ar'SO_2Cl{\cdot}AlCl_3 + C_6H_5{\cdot}C_6H_5 \longrightarrow \underset{\text{(III)}}{Ar'SO_2(AlCl_3)C_6H_4{\cdot}C_6H_5} \longrightarrow \text{Compound (II)}$$

Table 2 shows that only with benzenesulphonyl chloride was the desired product obtained.

TABLE 2

Sulphonyl chloride	Reaction time (days)	Monosulphone (III) Yield (%)	Monosulphone (III) M. p.	Disulphone (II) Yield (%)	Disulphone (II) M. p.
Ph	(*a*) 2	(*a*) 55	149·5—150° [4]	(*a*) 33	299—300° [2]
	(*b*) 7	(*b*) 39		(*b*) 60	
2,5-$C_6H_3Me_2$	(*a*) 2	(*a*) 72	124·5—125	(*a*) 0	
	(*b*) 7	(*b*) 98		(*b*) 0	
2,4,6-$C_6H_2Me_3$	(*a*) 2	(*a*) 50	115·5—116	(*a*) 0	
	(*b*) 7	(*b*) 98		(*b*) 0	

In this instance, the first-formed monosulphone (III; Ar′ = Ph) is slowly converted into the disulphone (II; Ar′ = Ph). The latter was not obtained by the action of benzenesulphonyl chloride and aluminium chloride on the monosulphone unless this was present as its oxonium complex with aluminium chloride. Presumably the monosulphone is sufficiently basic to take over aluminium chloride from the benzenesulphonyl chloride and hence to prevent sulphonylation. When both sulphone and sulphonyl chloride are present as their oxonium complexes, sulphonylation presumably proceeds through the small amount of free sulphone in the equilibrium mixture.

$$Ar{\cdot}Ar'SO_2 + AlCl_3 \rightleftharpoons Ar{\cdot}Ar'SO_2{\cdot}AlCl_3$$

The failure of *p*-xylene- and mesitylene-sulphonyl chlorides to effect disulphonylation of biphenyl provides a further example [1] of a reduction in the electrophilic character of the reagent, consequent on the inductive effect of *o*- and *p*-alkyl groups.

Experimental.—Preparation of sulphones.—The procedure was essentially that previously described by Holt and Pagdin,[1] except that in reactions involving biphenyl-4,4′-disulphonyl chloride [5] 2·5 moles of aluminium chloride per mole of the acid chloride were used. Similarly, in attempts to effect the disulphonylation of biphenyl, the latter was treated with two molecular proportions of sulphonylating agent.

The crude disulphones, after preliminary purification by shaking with benzene, in which they are virtually insoluble, were crystallised to constant m. p. from dioxan. When a mixture of the mono- and di-sulphones, described in Table 2, was shaken with benzene, the monosulphone dissolved readily and was recovered by evaporation of the filtered solution under reduced pressure. The crude material was crystallised to constant m. p. from benzene–light petroleum (b. p. 60—80°).

Purification of 4,4′-bis(biphenyl-4″-sulphonyl)biphenyl. To a stirred suspension of the crude sulphone (2 g.), m. p. 345—346°, in ethylene dichloride (50 ml.) was added freshly powdered aluminium chloride (2 g.). When the sulphone had dissolved, the deep red-purple solution was filtered, to remove undissolved aluminium chloride, and to it was added anisole (3 g.) in ethylene dichloride (50 ml.). The precipitated sulphone (1·95 g., 98%), m. p. 361—363° (decomp.), was separated, washed with ethylene dichloride–anisole (10 : 1), and finally washed with ethylene dichloride. Crystallisation from a large volume of chloroform failed to raise the m. p. of the sulphone (Found: C, 73·8; H, 4·5; S, 11·0. Calc. for $C_{36}H_{26}O_4S_2$: C, 73·8; H, 4·4; S, 10·9%).

The following new sulphones were prepared: 4,4′-*Di*-(p-*xylylsulphonyl*)*biphenyl* (Found: C, 68·6; H, 5·5; S, 13·0. $C_{28}H_{26}O_4S_2$ requires C, 68·5; H, 5·3; S, 13·1; 4,4′-*Di*(*mesitylsulphonyl*)*biphenyl* (Found: C, 69·2; H, 5·9; S, 12·3. $C_{30}H_{30}O_4S_2$ requires C, 69·5; H, 5·8; S, 12·4%); 4-p-*Xylylsulphonylbiphenyl* (Found: C, 74·9; H, 5·8; S, 9·9. $C_{20}H_{18}O_2S$ requires C, 74·5; H, 5·6; S, 9·9%); 4-*Mesitylsulphonylbiphenyl* (Found: C, 75·3; H, 6·0; S, 9·5. $C_{21}H_{20}O_2S$ requires C, 75·0; H, 5·95; S, 9·5%).

Microanalyses were carried out by Mr. B. Manohin.

THE MANCHESTER COLLEGE OF SCIENCE AND TECHNOLOGY,
MANCHESTER 1. [*Received, December 29th*, 1964.]

[4] R. Adams, W. Reifschneider, and M. D. Nair, *Croat. Chem. Acta*, 1957, **29**, 277 (*Chem. Abs.*, 1959, **53**, 16,146).
[5] J. Feldman, *Helv. Chim. Acta*, 1931, **14**, 764.

774. *The Reduction of* 1-*Chloro- and* 1,4-*Dichloro*-5-*nitronaphthalene*

By R. W. Dennison and F. Scheinmann

The reduction of 1-chloro- and 1,4-dichloro-5-nitronaphthalene was re-examined because, until recently, the purity of the starting materials was uncertain.[1] 1-Chloro-5-nitronaphthalene was readily reduced catalytically in the presence of Raney nickel. The analysis, spectral values, and derivatives of the major product (m. p. 71·5°) are consistent with those for 5-chloro-1-naphthylamine, but the melting point and that of its acetyl derivative are different from those previously reported.[2,3] Gas–liquid chromatography confirmed the purity of the product and showed that the only other product in the crude reaction mixture was a trace of 1-naphthylamine. By reduction of 1-chloro-5-nitronaphthalene with stannous chloride and hydrochloric acid, as described by Friedlander *et al.*,[2] we obtained a mixture of 5-chloro-1-naphthylamine, 5,8-dichloro-1-naphthylamine, and 1-naphthylamine. Reduction with neutral sodium dithionite of both 1-chloro- and 1,4-dichloro-5-nitronaphthalene yields the corresponding amines and also unidentified products containing sulphur.

Recently,[4] 1,4-dichloro-5-nitronaphthalene has been conveniently hydrogenated in the presence of Raney nickel to give the corresponding amine. Use of more active Raney nickel [5] led to reductive removal of chlorine to yield 5-chloro-1-naphthylamine with subsequent deactivation of the catalyst, which occurs concurrently with the formation of 5,8-dichloro-1-naphthylamine.

Experimental.—Catalytic hydrogenations were carried out with Raney nickel T-4 catalyst prepared according to the method of Nishimura.[5]

Reduction of 1-*chloro*-5-*nitronaphthalene.*—(a) *By atmospheric hydrogenation in presence of Raney nickel.* 1-Chloro-5-nitronaphthalene [1] (1·04 g., 0·005 mole) in benzene (125 ml.) was hydrogenated at atmospheric pressure in the presence of Raney nickel.[5] Anhydrous magnesium sulphate (1·0 g.) was also added to absorb water formed during the hydrogenation. Hydrogenation ceased after 6·5 hr. (uptake 361 ml., theory requires 367 ml. at 25°). Removal of the residue and evaporation of the solvent from the filtrate gave a pale yellow solid (0·9 g.), m. p. 68—69°. Analysis by gas–liquid chromatography showed that the reaction product contained a trace of 1-naphthylamine. Crystallisation from light petroleum (b. p. 60—80°) gave chromatographically pure 5-*chloro*-1-*naphthylamine* (0·65 g.) as white plates, m. p. 71·5° (lit.,[2,3] 85°) (Found: C, 66·9; H, 4·3; Cl, 19·6; N, 7·6. $C_{10}H_8ClN$ requires C, 67·2; H, 4·5; Cl, 19·9. N, 7·8%.) The nuclear magnetic resonance (n.m.r.) spectrum shows a broad peak at 6·1 τ (−NH_2) and a complex group of aromatic absorptions from 2·3 to 3·4 τ, the ratio of the intensities of the groups being 1 : 3.

The N-*acetyl derivative* crystallised from aqueous ethanol as white needles, m. p. 202° (lit.,[2] 128°) (Found: Cl, 16·2; N, 6·4. $C_{12}H_{10}ClNO$; requires Cl, 16·1; N, 6·4%). The n.m.r. spectrum shows peaks at 7·68 τ ($CH_3CO{\cdot}N$−), 1·82 τ (NH·CO), and aromatic resonances from 2·1 to 2·6 τ.

The N-*benzoyl derivative* crystallised from aqueous ethanol as white needles, m. p. 173—174° (Found: Cl, 12·5; N, 4·9. $C_{17}H_{12}ClNO$ requires Cl, 12·6; N, 5·0%).

(b) *By catalytic hydrogenation.* 1-Chloro-5-nitronaphthalene (1·04 g.) in benzene (75 ml.) was hydrogenated at 4 atm. in the presence of Raney nickel [5] and anhydrous magnesium sulphate (1·0 g.). When the pressure was constant (3·41 atm.; theory 3·39 atm. at 25° in a 700-ml. reaction vessel), the crude product was isolated (1·0 g.) and shown to contain a trace (1%) of 1-naphthylamine. Crystallisation from light petroleum (b. p. 60—80°) yielded 5-chloro-1-naphthylamine (m. p. 71·5°), which was shown by mixed m. p., gas–liquid chromatography, and spectroscopy to be identical with the compound obtained previously.

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[2] P. Friedlander, S. Karamessinis, and O. Schenk, *Chem. Ber.*, 1922, **55***B*, 45.
[3] H. F. Bassilios, *Bull. Soc. chim. France*, 1951, 651.
[4] A. P. Lurie, G. H. Brown, J. R. Thirtle, and A. Weissberger, *J. Amer. Chem. Soc.*, 1961, **83**, 5015.
[5] S. Nishimura, *Bull. Chem. Soc. Japan*, 1959, **32**, 61.

(c) *By stannous chloride and hydrochloric acid.* 1-Chloro-5-nitronaphthalene (2·0 g.) in ethanol (20 ml.) was refluxed with stannous chloride (10 g.) and hydrochloric acid (36%, 20 ml.) for 45 min. The mixture was cooled and the resulting white precipitate was collected and washed with ethanol. The combined filtrate was made alkaline with aqueous sodium hydoxide (4N; 85 ml.) and extracted with ether. Removal of the solvent and analysis of the oily product (0·31 g.) by gas–liquid chromatography showed a mixture of 1-naphthylamine and 5-chloro- and 5,8-dichloro-1-naphthylamine.

The white precipitate was dissolved in water (200 ml.) and made alkaline with aqueous sodium hydroxide to yield 5-chloro-1-naphthylamine (0·79 g.).

(d) *By sodium dithionite.* A solution of 1-chloro-5-nitronaphthalene (2·5 g.) and sodium dithionite (12·5 g.) in ethanol (50 ml.) and water (40 ml.) was boiled for 40 min. The mixture was poured into water (200 ml.) and the precipitate was extracted with ether. Removal of the solvent from the organic portion gave 5-chloro-1-naphthylamine (18%), m. p. 71°, undepressed on admixture with the sample prepared previously.

Evaporation of the aqueous extract to half its volume precipitated a white solid (1·0 g.), which was collected and dried. Recrystallisation from water gave a *sodium salt*, as white needles, soluble in mineral acid but reprecipitated by aqueous sodium hydroxide (Found: Cl, 12·7; N, 4·9; S, 11·4. $C_{10}H_7ClNNaO_3S$ requires Cl, 12·7; N, 5·0; S, 11·4%).

Reduction of 1,4-dichloro-5-nitronaphthalene. (a) *By catalytic hydrogenation.* Attempts to hydrogenate 1,4-dichloro-5-nitronaphthalene in the presence of Raney nickel [5] under the same conditions and molar scale as described for the hydrogenations of 1-chloro-5-nitronaphthalene were unsuccessful. Hydrogenation was slow and incomplete, despite further additions of Raney nickel (after 8½ hr. 182 ml. of hydrogen absorbed). Analysis by thin-layer and gas–liquid chromatography showed that 5-chloro-1-naphthylamine, 5,8-dichloro-1-naphthylamine, and 1,4-dichloro-5-nitronaphthalene were present in the mixture.

(b) *By sodium dithionite.* 1,4-Dichloro-5-nitronaphthalene (2·5 g.) in ethanol (50 ml.) was boiled under reflux with sodium dithionite (12·5 g.) in water (40 ml.) for 40 min. The mixture was poured into water (200 ml.) and the residue extracted with ether. Evaporation of the solvent from the organic portion gave a lilac solid (0·89 g.), m. p. 102—103°. Recrystallisation from aqueous ethanol or petroleum ether (b. p. 80—100°) gave 5,8-dichloro-1-naphthylamine as white needles, m. p. 104° [2] (lit.,[4] 102—103°) (Found: C, 56·3; H, 3·0; N, 6·4. Calc. for $C_{10}H_7Cl_2N$: C, 56·6; H, 3·3; N, 6·6%).

Reduction in volume of the aqueous portion gave a *sodium salt* (Found: N, 4·3; S, 97; ($C_{10}H_6Cl_2NNaO_3S$ requires N, 4·4; S, 10·2%). *N*-Acetyl-5,8-dichloro-1-naphthylamine recrystallised from ethanol as white needles, m. p. 210° (lit.,[2] 202°) (Found: Cl, 29·0; N, 5·5. Calc. for $C_{12}H_9Cl_2NO$: Cl, 28·0; N, 5·5%).

N-Benzoyl-5,8-dichloro-1-naphthylamine recrystallised from ethanol as white needles, m. p. 180° (Found: Cl, 21·9; N, 4·3. $C_{17}H_{11}Cl_2NO$ requires Cl, 21·9; N, 4·3%).

Analysis by chromatography. It was necessary to analyse reaction mixtures by thin-layer and gas–liquid chromatography. Thus, while 5-chloro-1-naphthylamine could not be separated from 1-naphthylamine by thin-layer chromatography on bound alumina, separation by gas–liquid chromatography on 10% Apiezon L was satisfactory. On the other hand, 1-chloro-5-nitronaphthalene and 5-chloro-1-naphthylamine, which have identical retention times in gas–liquid chromatography, were separated by thin-layer chromatography.

Bound aluminium oxide G (Merck) thin-layer chromatography plates were activated at 120° for 1 hr. before use. A 5% solution of benzene in light petroleum (b. p. 80—100°) separated the chloronitronaphthalenes but, for the chloronaphthylamines, the plates were developed with a 15% solution and viewed in ultraviolet light.

The gas–liquid chromatography was carried out with the Pye Argon Chromatograph under operating conditions similar to those for the chloronitronaphthalenes.[1] Retention times relative to 1-nitronaphthalene are given in brackets: 1-naphthylamine (0·8), 5-chloro-1-naphthylamine (1·9), 5,8-dichloro-1-naphthylamine (3·2).

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775. *Crystallographic Data for the 2′-Halogenobiphenyl-4-carboxylic Acids and 4-Acetyl-2′-halogenobiphenyls*

By G. W. Gray, H. H. Sutherland, and D. W. Young

In connection with recent studies of the liquid crystalline properties of 2-substituted derivatives of biphenyl,[1] a detailed knowledge of the crystal structures of 2-substituted biphenyls is of immediate interest. With this aim in mind, a preliminary crystallographic investigation of the 2′-halogenobiphenyl-4-carboxylic acids and 4-acetyl-2′-halogenobiphenyls has been made.

The unit-cell dimensions were obtained photographically (Weissenberg and precession goniometers) using Cu-K_α and Mo-K_α radiations, and the densities were measured by flotation using aqueous cadmium *n*-dodecatungstaborate or potassium iodide.

Work is proceeding on determining the structure of 4-acetyl-2′-fluorobiphenyl, and investigation has begun on the structures of 4-acetyl-2′-chlorobiphenyl and the probably isomorphous pair—2′-fluoro- and -chloro-biphenyl-4-carboxylic acids.

Crystal data

Unit-cell dimensions	Space group	U (Å^3)	M	Z	D_c	D_m	M. p.
4-Acetyl-2′-halogenobiphenyls							
F $a = 13{\cdot}69$; $b = 5{\cdot}97$; $c = 14{\cdot}77$ Å; $\beta = 116° 10'$	$P2_1/c$	1083·1	214·2	4	1·31	1·31	85·5—86·5°
Cl $a = 4{\cdot}00$; $b = 38{\cdot}51$; $c = 7{\cdot}52$ Å; $\beta = 100° 4'$	$P2_1/c$	1139·6	230·7	4	1·34	1·36	55·5
Br $a = 23{\cdot}43$; $b = 12{\cdot}70$; $c = 8{\cdot}07$ Å	$Pbca$	2401·3	275·1	8	1·52	1·53	81—82
I $a = 23{\cdot}91$; $b = 12{\cdot}49$; $c = 8{\cdot}45$ Å	$Pbca$	2523·5	322·2	8	1·70	1·71	86—87
2′-Halogenobiphenyl-4-carboxylic acids							
F $a = 3{\cdot}86$; $b = 34{\cdot}35$; $c = 7{\cdot}78$ Å; $\beta = 102° 20'$	$P2_1/c$	1007·5	216·2	4	1·43	1·44	232·5
Cl $a = 3{\cdot}94$; $b = 35{\cdot}81$; $c = 7{\cdot}61$ Å; $\beta = 101° 6'$	$P2_1/c$	1053·4	232·7	4	1·47	1·46	252
Br $a = 4{\cdot}01$; $b = 35{\cdot}81$; $c = 7{\cdot}97$ Å	$P2_1/m$ or $P2_1$	1075·7	277·1	4	1·71	1·70	242·5
I $a = 4{\cdot}15$; $b = 7{\cdot}73$; $c = 17{\cdot}79$ Å; $\alpha = 99° 4'$; $\beta = 91° 23'$; $\gamma = 97° 34'$	$P\bar{1}$ or $P1$	566·3	324·1	2	1·90	1·90	241

Experimental.—Materials. The ketones were prepared by Friedel–Crafts acylation of the 2-halogenobiphenyls, and oxidised to the 2′-halogenobiphenyl-4-carboxylic acids by sodium hypobromite in 50% aqueous dioxan. Full details [2] of these and related preparations will be published in the near future. The products were crystallised from ethanol and from petroleum (b. p. 40—60°) until the m. p.s (see Table) were constant. The results [2] from combustion analyses were satisfactory, and the products shown to be free from isomers by gas–liquid chromatography carried out on the ketones and on samples of the methyl esters prepared from the acids.

Suitable crystals of the ketones and the 2′-fluoro- and 2′-chloro-biphenyl-4-carboxylic acids were obtained by using ethanol as solvent. In the case of the 2′-bromo- and -iodo-biphenyl-4-carboxylic acids, the preferred solvents were ethyl acetate or toluene. Small crystals were often the best for the crystallographic studies, even moderately sized crystals being frequently twinned.

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[*Received, November 2nd*, 1964.]

[1] S. J. Branch, D. J. Byron, G. W. Gray, A. Ibbotson, and B. M. Worrall, *J.*, 1964, 3279, and preceding Parts.

[2] D. J. Byron, G. W. Gray, and R. C. Wilson, unpublished work.