The yield of the former was about two-thirds that of the latter.

The product melted at 110–111°; $[\alpha]_D^{28}$ −8.5° (*c*, 1.04 in methanol).

N^{ϵ}-Tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine. Saponification of N^{α}-carbobenzyloxy-N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ester (either methyl or ethyl) by shaking it with *N* sodium hydroxide, acidification of the aqueous solution and extraction with ethyl acetate yielded an oil from which no crystals could be obtained.

Hydrogenation of this oil in 15% solution of glacial acetic acid in anhydrous methanol, with palladium as a catalyst, yielded a glossy residue which was not readily soluble in water. Addition of absolute ethanol resulted in formation of crisp, white microcrystals, m.p. 167–168°.

The product was ninhydrin-positive and gave a single spot on a paper chromatogram developed either with the Partridge solvent system (R_f 0.90) or with the 2-butanol–ammonia (3:1) system (R_f 0.65). The sterical configuration of the product was confirmed by the leucine aminopeptidase digestion which completely hydrolyzed the dipeptide leaving only N^{ϵ}-tosyl-L-lysine in the digest. The elementary composition of the product was also confirmed.

Anal. Calcd. for $C_{26}H_{38}O_7N_4S_2$: C, 53.28; H, 6.57; N, 9.61. Found: C, 52.92; H, 6.63; N, 9.72.

Carbobenzyloxyglycyl-N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester. N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester hydrochloride was prepared from its N^{α}-carbobenzyloxy derivative by a catalytic hydrogenation in methanol in the presence of equimolecular amount of *N* hydrochloric acid, with palladium as a catalyst. An oil was obtained in a virtually quantitative yield, which gave a single ninhydrin spot in either of the above-described solvent systems. Leucine aminopeptidase hydrolysis of the oil gave a single ninhydrin spot of N^{ϵ}-tosyl-L-lysine.

This oil was coupled with an equimolecular amount of carbobenzyloxyglycine by the carbodiimide method; *i.e.*, N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester hydrochloride (1.25 g.), carbobenzyloxyglycine (0.41 g.), triethylamin (0.30 ml.) and dicyclohexylcarbodiimide (0.41 g.) were dissolved in dioxane (20 ml.). The mixture was left to stand overnight at room temperature. A precipitate formed which was removed by filtration (0.40 g., corresponds to 2.05 mmoles of dicyclohexylurea). The filtrate was evaporated *in vacuo* to dryness (40° bath temp.) and the oily residue dissolved in ethyl acetate. This solution was purified in the usual way (washed consecutively with 2*N* hydrochloric acid, *N* sodium bicarbonate, followed with distilled water until the washings were neutral), then dried over magnesium sulfate, and evaporated *in vacuo* to dryness. An oily residue (1.15 g.) was obtained. This residue was dissolved in methanol and water was added until permanently turbid. On standing overnight crystals appeared. Slow addition of water yielded more crystals. Both crops (0.82 g., 53% yield) had a melting point of 121–124°. Recrystallization of an aliquot increased the m.p. to 125.5–126°; its optical rotation was $[\alpha]_D^{23}$ −12.6° (*c*, 1.06 in ethanol).

Anal. Calcd. for $C_{38}H_{51}O_{10}N_5S_2$: C, 56.90; H, 6.41; N, 8.73; S, 7.99. Found: C, 56.77; H, 6.29; N, 8.80; S, 8.00.

Catalytic hydrogenation of the aliquot produced in virtually a stoichiometric yield an oil, R_f 0.91 (Partridge), 0.85 (2-butanol–ammonia), completely digestible by leucine amino peptidase, amino acid ratios in digest gly$_{1.0}$$\epsilon$-tos-lys$_{1.9}$.

Dicarbobenzyloxy-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester was prepared from dicarbobenzyloxy-L-lysine and the ethyl ester of N^{ϵ}-tosyl-L-lysine hydrochloride following the procedure described above for the preparation of N^{α}-carbobenzyloxy-N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester. On addition of water to the reaction mixture an oil was obtained. Trituration of this oil with 5% aqueous sodium bicarbonate resulted in a formation of crystals. These were washed twice with water, filtered, and dried *in vacuo* over phosphorus pentoxide; 72.4% yield, m.p. 115–118°. Recrystallization from methanol and water increased the m.p. to 121–122° $[\alpha]_D^{27.5}$ −11.3° (*c*, 2.47 in methanol).

Anal. Calcd. for $C_{37}H_{48}O_9N_4S$: C, 61.30; H, 6.67; S, 4.42. Found: C, 61.19; H, 6.85; S, 4.33.

N^{α}-Carbobenzyloxy-N^{ϵ}-tosyl-L-lysyl-L-valine methyl ester was prepared from N^{α}-carbobenzyloxy-N^{ϵ}-tosyl-L-lysine (2.27 g.) and the methyl ester of L-valine hydrochloride (1.05 g.) following the procedure described above for N^{α}-carbobenzyloxy-N^{ϵ}-tosyl-L-lysyl-N^{ϵ}-tosyl-L-lysine ethyl ester. The yield (1.81 g.) was 63.4%; m.p. 97.5–98.5°, on recrystallization from methanol-water raised to 99.5–100°; $[\alpha]_D^{26}$ −14.3° (*c*, 2.51 in ethanol).

Anal. Calcd. for $C_{27}H_{37}O_7N_3S$: C, 59.21; H, 6.81; S, 5.85. Found: C, 59.14; H, 6.64; S, 5.84.

Catalytic hydrogenation of a small aliquot resulted in an almost quantitative yield of decarbobenzoxylated dipeptide, R_f = 0.89 (Partridge), single spot in (2-butanol–ammonia), completely digestible by LAP, amino acid ratios in digest ϵ-tos-lys$_1$val$_1$.

Acknowledgment. Gratitude is expressed to Dr. K. Hofmann for numerous discussions and to Mrs. E. T. Schwartz for the chromatographic and leucine aminopeptidase-digestion assays.

OTTAWA, CANADA

[CONTRIBUTION FROM THE RESEARCH AND DEVELOPMENT DEPARTMENT, COLGATE-PALMOLIVE CO.]

Distribution of *para* and *ortho* Isomers in Some Model Long Chain Alkylbenzenesulfonates

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A series of long chain 1-phenyl- and 2-phenylalkanes has been prepared and sulfonated. The sulfonate distribution was about 80% *para*, 15% *ortho* for the 1-phenylalkanes and about 90% *para*, 7% *ortho* for the secondary 2-phenylalkanes. The isomers were characterized by conversion to their *S*-benzylisothiuronium salts and by infrared spectra. Some physical properties of the alkylbenzenesulfonates are described.

The preparation and properties, including surface activity, of some isomeric sodium alkylbenzenesulfonates were described in a previous publication.[1b] The position of the sulfonate group in the

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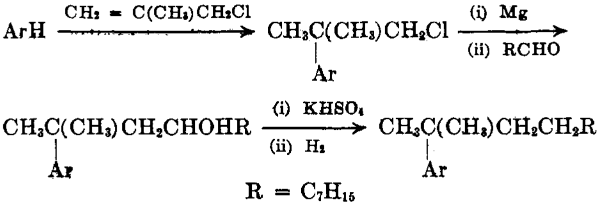
(1b) F. W. Gray, J. F. Gerecht, and I. J. Krems, *J. Org. Chem.*, 20, 511 (1955).

benzene ring, the location of the phenyl group and the length of the alkyl chain were factors shown to affect the surface active properties of these compounds. The present work extends our study of model primary and secondary isomeric alkylbenzenesulfonates and also includes an examination of model tertiary alkylbenzenesulfonate.

The method of synthesis used for the primary and secondary alkylbenzenes was the same as previously reported. To synthesize the model tertiary alkylbenzene, 2-phenyl-2-methylundecane, two routes were investigated. In the first, α-methylstyrene was treated with hydrogen chloride to form α-chlorocumene. This intermediate was condensed with nonylmagnesium bromide and a 30% yield of crude product was obtained. When this material was distilled through a Podbielniak column, fractions having a wide boiling range were obtained and the indexes of refraction varied considerably. The purity of the alkylbenzene was not satisfactory for our purpose.

In the second route, the method of Weinmayr was used.[2] Benzene was alkylated with methallyl chloride to form neophyl chloride. The Grignard reagent prepared from this compound was treated with octanal. The resultant carbinol, 2-phenyl-2-methyl-4-undecanol, was isolated in a 76% yield. The alcohol was dehydrated and reduced to 2-phenyl-2-methylundecane.



$$ArH \xrightarrow{CH_2 = C(CH_3)CH_2Cl} \underset{\displaystyle Ar}{CH_3C}(CH_3)CH_2Cl \xrightarrow[\text{(ii) } RCHO]{\text{(i) } Mg}$$

$$\underset{\displaystyle Ar}{CH_3C}(CH_3)CH_2CHOHR \xrightarrow[\text{(ii) } H_2]{\text{(i) } KHSO_4} \underset{\displaystyle Ar}{CH_3C}(CH_3)CH_2CH_2R$$

$$R = C_7H_{15}$$

Under the sulfonation conditions used, the primary and secondary hydrocarbons were sulfonated in about 98% yields. The sulfonate isomer distribution for the 1-phenylalkanes was about 80% *para* and 15% *ortho;* whereas for the 2-phenylalkanes, the sulfonate content was about 90% *para* and 7% *ortho*. The *para* isomers in both series were white crystalline non-tacky compounds. The *ortho* sulfonates in the 1-phenyl series were slightly yellow in color, were less crystalline than the *para* isomers and they were hygroscopic and tacky. The *ortho* sulfonates in the 2-phenyl series were light brown in color and at room temperature were viscous tacky pastes.

The sulfonation of the model tertiary compound, 2-phenyl-2-methylundecane, with 20% oleum offered no difficulty and proceeded in the same way as the primary and secondary alkylbenzenes. The neutralized white crystalline product, sodium (1,1-dimethyldecyl)benzenesulfonate, possessed strong infrared absorption bands at 12.00 μ and 13.12 μ. These same absorption bands are present in mixtures of *para* and *ortho* secondary alkylbenzenesulfonates. On recrystallization of such a mixture, the *para* sulfonate isomer (12.00 μ) separates as a crystalline compound whereas the *ortho* sulfonate isomer (13.12 μ) is amorphous and difficult to obtain in a purified state. Attempts to accomplish a similar separation with the sulfonation product of 2-phenyl-2-methylundecane were not successful. After repeated recrystallizations, the 12.00 μ and 13.12 μ bands remained unchanged. A phase solubility analysis was undertaken to determine the homogeneity of the compound. The results unambiguously demonstrated the product to be essentially a single compound. It appears that in the isolated compound, sodium (1,1-dimethyldecyl)benzenesulfonate, the tertiary structure of the alkylbenzene and not the presence of *ortho* sulfonate substitution is responsible for the 13.12 μ absorption band.

A detailed discussion of the sodium alkylbenzenesulfonates in conjunction with their wetting, detergency, and foaming properties and a comparison of their behavior in these respects to commercial sodium (tetrapropylene)benzenesulfonate is to appear in another publication. In respect to sulfonate isomerism, the results obtained support the observation previously reported[1b] that the *para* sulfonates are superior in detergency but inferior in wetting action to their respective *ortho* isomers. In regard to foaming, however, it is now found that *para* rather than *ortho* substitution tends generally to favor better foaming action. This conclusion corrects the statement made in the earlier paper, based on relatively few experiments, that the *ortho* orientation is the favored position for foam height.

The extreme insolubility of sodium *p*-dodecylbenzenesulfonate places a restriction on the study of the surface active behavior of primary alkylbenzenesulfonates. Sodium *p*-octylbenzenesulfonate and sodium *p*-decylbenzenesulfonate have better solubility properties but the surface activity properties examined are not as good as for the higher molecular weight compound. When the phenyl group is shifted from the 1- to the 2-position in an alkylbenzenesulfonate, an appreciable increase in solubility occurs in the isomer. Consequently, higher molecular weight sulfonates may be utilized for detersive studies in the secondary series than was found possible for the primary alkylbenzenesulfonates.

The foaming ability of the *para* sulfonates of the secondary 2-phenylalkanes is dependent upon chain length as well as concentration. The foam height obtained for sodium *p*-(1-methyldodecyl)-benzenesulfonate is greater than obtained for sodium (tetrapropylene)benzenesulfonate but the height of foam obtained with sodium *p*-(1-methylhexadecyl)benzenesulfonate is less than for the commercial product at all concentrations ex-

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(2) V. Weinmayr, U. S. Patent 2,467,170 (1949).

amined (0.015–0.045%). In detergency, however, sodium *p*-(1-methyldodecyl)-, sodium *p*-(1-methyltetradecyl)-, and sodium *p*-(1-methylhexadecyl)-benzenesulfonate are superior to commercial sodium (tetrapropylene)benzenesulfonate. The tertiary compound, sodium *p*-(1,1-dimethyldecyl)benzenesulfonate, has overall surface active properties that closely resemble commercial sodium (tetrapropylene)benzenesulfonate and its wetting ability is superior to the sodium *p*-sulfonates of the 1-phenylalkanes and 2-phenylalkanes.

EXPERIMENTAL

Phenylalkanes. 1-Phenyloctane, 1-phenyldecane, and 1-phenyldodecane were prepared by condensing the acid chlorides of octanoic, decanoic, and dodecanoic acids with benzene under Friedel Crafts conditions followed by reduction of the resulting ketones to hydrocarbons by the Wolff-Kishner reaction. 2-Phenylundecane, 2-phenyltridecane, 2-phenylpentadecane, and 2-phenylheptadecane were prepared by conversion of the appropriate fatty acids to acid chlorides and then to the phenones by using the Friedel Crafts reaction. The phenyl ketones thus obtained were treated with Grignard reagent and hydrolyzed. The tertiary alcohols formed were dehydrated and the phenylalkenes produced were hydrogenated to the desired products.[1b]

2-Phenyl-2-methylundecane was prepared by the general procedure described by Weinmayr.[2] Neophylmagnesium chloride was prepared from 75.5 g. of neophyl chloride[3] and 12.0 g. of magnesium in 425 ml. of ethyl ether. The chloride was added in a period of 3 hr., and the mixture refluxed for an additional 2 hr. A solution of 50 g. of octanal in 75 ml. of ethyl ether was then added in 35 min., the mixture refluxed for 2 hr., and then allowed to stand overnight at room temperature. The reaction mixture was poured into 500 g. of cold water and acidified with hydrochloric acid. The ether layer was washed with 5% sodium bicarbonate solution and the solvent removed by distillation. The residue was heated for 2 hr. at 200–290° in the presence of potassium acid sulfate. The reaction mixture was filtered and 64 g. of material, presumably a mixture of 2-phenyl-2-methyl-3-undecene and 2-phenyl-2-methyl-4-undecene, was obtained upon distillation. To a 40-g. fraction (b.p. 111°/1 mm.) was added 200 g. of glacial acetic acid and 0.4 g. of 5% palladium on charcoal catalyst. The reaction mixture was placed in a Parr apparatus and the theoretical amount of hydrogen was absorbed within 4 hr. at room temperature and 1–3 atm. pressure. The catalyst was removed by filtration and the acetic acid by distillation. At 128–130° (1.5–2.0 mm.) there was obtained 35.5 g. of hydrocarbon. This material was washed three times with 96% sulfuric acid, then with sodium bicarbonate. The purified product was re-distilled at 123° (1 mm.) to yield 27.2 g. The physical properties of the alkylbenzenes used for sulfonation are listed in Table I.

TABLE I

PHYSICAL PROPERTIES OF ALKYLBENZENES

Hydrocarbons	B.P.	Mm.	d_4^{25}	n_D^{25}
1-Phenyloctane	87–88	1	0.8541	1.4825
1-Phenyldecane	112	1	0.8520	1.4812
1-Phenyldodecane	148	1	...	1.4818[a]
2-Phenylundecane	92–94	0.2	0.8507	1.4804
2-Phenyltridecane	132–133	0.8	0.8612	1.4794
2-Phenylpentadecane	192	5	0.8577	1.4796
2-Phenylheptadecane	156–158	0.8	0.8522	1.4782
2-Phenyl-2-methyl-undecane	123	1	0.8599	1.4848

[a] Refractive index taken at 20.

2-Phenyl-2-methyl-4-undecanol. The neophylmagnesium chloride and octanal condensation product prepared in the manner described in the preparation of 2-phenyl-2-methylundecane was hydrolyzed in cold dilute aqueous acid solution. The reaction product was extracted with ethyl ether and dried over magnesium sulfate. The product distilled in 76% yield at 150–155° (1.5 mm.); d_4^{25} 0.9234; n_D^{25} 1.5010.

Sulfonation of phenylalkanes. The 1-phenylalkanes and 2-phenylalkanes were sulfonated in about 98% yields with 20% oleum, neutralized with sodium hydroxide, ether extracted, inorganic salt removed, and the *ortho-para* isomers isolated under conditions essentially the same as previously described for 1-phenyloctane and 2-phenyltridecane.[1b] The *para* and *ortho* sulfonate yields are given in Table II.

TABLE II

YIELD AND DISTRIBUTION OF SULFONATE ISOMERS

Alkylbenzene	Sulfonation Yield, %[a]	Isomer Distribution *para*, %	*ortho*, %
1-Phenyloctane	98	81	15
1-Phenyldecane	99	80	15
1-Phenyldodecane	98	81	14
2-Phenylundecane	99	86	7
2-Phenyltridecane	98	91	6
2-Phenylpentadecane	98	90	7
2-Phenylheptadecane	98	89	7

[a] Yield based upon recovered unsulfonated material.

The isomers were identified by derivative formation and by infrared spectral data. The *para* sulfonates of the primary alkylbenzenes exhibit a doublet at 11.90, 12.28 μ while the *ortho* sulfonates exhibit a single absorption band at 13.36 μ. These characteristic isomer peaks were established from the properties of isomeric sodium dodecylbenzenesulfonates that were prepared by an unequivocal method of synthesis. The *para* sulfonates of the secondary alkylbenzenes possess a single *para* absorption at about 12.00 μ whereas the *ortho* sulfonates have a strong absorption at about 13.12 μ. Conceivably, an isomerization of secondary to tertiary structure during sulfonation could account for the small amount of sulfonate having an absorption band at 13.12 μ. However, the physical characteristics and the infrared spectrum of the sulfonation product obtained from the tertiary compound, 2-phenyl-2-methylundecane, do not support the possibility that such an isomerization occurs. The melting points and elemental analyses for the *S*-benzylisothiuronium salts of isomeric primary and secondary alkylbenzenesulfonates are given in Table III.

Sulfonation of 2-phenyl-2-methylundecane with 20% oleum offered no difficulty and presented the same appearance as the acid mixes of the primary and secondary alkylbenzenes. The reaction mixture was neutralized with sodium hydroxide, dried, and extracted with anhydrous ethyl ether. The ether soluble material was transferred to a separatory funnel and partitioned between equal volumes of water and ethyl ether. The ether layer contained less than 1% unsulfonated material while the water layer, which was recovered by evaporation to dryness, was found to contain an 80% yield of sodium (1,1-dimethyldecyl)benzenesulfonate. A recrystallization of 10.0 g. of the product from 100 ml. of water gave 7.8 g. of a crystalline, white and odorless compound (I). Its infrared spectrum suggested an *ortho-para* isomer mixture since absorption peaks were present at 13.12 (*ortho*) and 12.00 (*para*) μ. However, repeated recrystallizations and extraction procedures did not cause the 13.12 μ absorption peak to disappear. To establish the degree

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(3) W. T. Smith, Jr., and J. T. Sellas, *Org. Synthesis*, **32**, 90 (1952).

TABLE III

S-Benzylisothiuronium Salts of Isomeric Alkylbenzenesulfonates

$RC(R')HC_6H_4SO_3Na$				M.P.	Nitrogen, %		Sulfur, %	
Orientation	R	R′	M.P.[a]	(Ref. 1b)	Calcd.	Found	Calcd.	Found
p	C_7H_{15}	H	125.6–125.8	125.6–126.0	...	...	...	...
o	C_7H_{15}	H	98–99[b]	105.5–106	...	...	...	...
p	C_9H_{19}	H	94.2–95.3	...	6.03	6.20	13.78	13.23
o	C_9H_{19}	H	113–114	...	6.03	5.94	13.78	13.95
p	$C_{11}H_{23}$	H	117–118	117–118	...	...	...	...
o	$C_{11}H_{23}$	H	101–103	101–103	...	...	...	...
p	C_9H_{19}	CH_3	104.0–104.4	...	5.85	5.66	13.37	13.98
o	C_9H_{19}	CH_3	108.6–109	...	5.85	5.72	13.37	13.46
p	$C_{11}H_{23}$	CH_3	105.9–106.2	105–106	5.53	5.43	12.63	12.93
o	$C_{11}H_{23}$	CH_3	86–86.5	...	5.53	5.38	12.63	12.90
p	$C_{13}H_{27}$	CH_3	106–106.9	...	5.24	5.06	11.97	12.05
o	$C_{13}H_{27}$	CH_3	113.7–115	...	5.24	5.15	11.97	12.07
p	$C_{15}H_{31}$	CH_3	109–109.4[c]	...	4.98	4.91	11.38	11.48
o	$C_{15}H_{31}$	CH_3	92.2–93.2	...	4.98	4.60	11.38	11.24

[a] All melting points were taken after drying at least six hours over phosphorus pentoxide at 78 (2 mm.). [b] On resolidification of the melt, the melting point was 106–106.8. [c] *p*-Toluidine derivative had a melting point of 119.1–119.5. *Anal.* Calcd. for $C_{30}H_{49}NO_3S$: N, 2.78; S, 6.35. Found: N, 2.73; S, 6.36.

of purity a phase solubility analysis[4] was conducted with I. The results of the analysis show conclusively that the compound isolated is essentially a single component with a 5.5% solubility in water at 46.5°. The *S*-benzylisothiuronium derivative of I, which is presumably the *para* isomer, melted at 126–128°.

Anal. Calcd. for $C_{26}H_{40}N_2O_3S_2$: N, 5.60; S, 13.62. Found: N, 5.63; S, 13.40.

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[4] W. J. Mader, *Organic Analysis*, Interscience Publishers Inc., New York, Vol. II., 253 (1954).

[Contribution from the Benzol Products Co.]

Isomer Distribution of Some Chloromethylated Alkylbenzenes

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Received May 9, 1960

Some alkylbenzenes were chloromethylated and the isomer contents of the resulting products examined by infrared spectrometry and gas-liquid chromatography. *meta*-Substitution occurred and in increasing amounts for ethyl-, methyl-, *i*-propy, and *t*-butylbenzenes.

Ingold,[1] in commenting upon the data of Le Févre,[2] stated that it was inconceivable that no *meta* compounds were formed when ethyl- and *i*-propylbenzenes were mononitrated. It was later found[3] that not inconsiderable quantities of the *meta* isomers actually did result and that there was increasing *meta* substitution in the series methyl-, ethyl-, *i*-propyl-, *t*-butylbenzenes. H. C. Brown[4] recently reported analogous behavior for acetylation and benzoylation.

Nazarov and Semenovskii[6] determined the isomer distribution of chloromethylated ethyl-, *i*-propyl-, and *t*-butylbenzenes by oxidation to the corresponding dicarboxylic acids. No *meta* substituted compounds were recovered, but it is apparent that small quantities could have escaped detection by their semiquantitative oxidation technique. We decided to investigate the isomeric products of the chloromethylation reaction by hydrogenolysis and subsequent examination by means

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[1] C. K. Ingold, *Structure and Mechanism in Organic Chemistry*, Cornell University Press, Ithaca, New York, 1953, p. 257.

[2] R. J. W. Le Févre, *J. Chem. Soc.*, 1501 (1934).

[3] H. C. Brown and W. H. Bonner, *J. Am. Chem. Soc.*, **76**, 605 (1954).

[4] H. C. Brown and G. Marino, *J. Am. Chem. Soc.*, **81**, 5611 (1959).

[6] I. N. Nazarov and A. V. Semenovskii, *Izvest. Akad. Nauk. S.S.S.R. Otdel. Khim. Nauk.*, 100–111 (1957) [*Chem. Abstr.*, **51**, 10400 (1957)].