

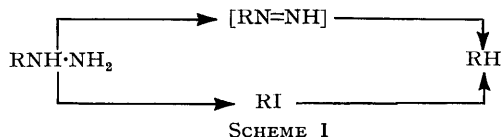
Oxidation of Alkylhydrazines by Iodine

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When treated with iodine in the presence of base, alkylhydrazines usually afford mixtures of the corresponding alkanes, alkenes, alkyl iodides, and (in aqueous media) alcohols, in relative amounts dependent upon the structure of the alkylhydrazine, the solvent, and the base used.

MONOALKYLHYDRAZINES, like hydrazine itself, are readily oxidised by a variety of reagents, the products varying with the oxidant used.¹ For example, when oxidation is effected by either hexacyanoferrate(III)² or periodate³ in the presence of base, an alkylhydrazine is converted smoothly into the corresponding alkane, probably³ *via* a carbanion derived from an intermediate alkyl-di-imide. An alternative procedure for effecting the same overall result involves oxidation of the alkylhydrazine by iodine in the presence of base to give the corresponding alkyl iodide, which may then be reduced (Scheme 1).



Both these procedures have been successfully applied to the synthesis of deoxy-sugars.⁴ At the outset of our work in this area, however, little was known about the effect of iodine on alkylhydrazines, and a detailed investigation of this type of reaction was therefore undertaken. We now present some of the results of this investigation.⁵

¹ P. A. S. Smith, 'Chemistry of Open-chain Nitrogen Compounds,' Vol. 2, Benjamin, New York, 1966, ch. 9.

² N. Kishner, *Russ. J. Phys. Chem.*, 1899, **31**, 1033.

RESULTS AND DISCUSSION

When treated with iodine in the presence of base, alkylhydrazines usually afford mixtures of products, the composition of which depends upon a number of factors, the most important being the solvent, the structure of the alkylhydrazine, and the base used.

(i) *Oxidations in Non-aqueous Media.*—Table 1 shows that oxidation of a given alkylhydrazine with iodine in non-aqueous media usually results in a mixture of the corresponding alkane, alkene, and alkyl iodide. The alkane is presumably formed *via* an intermediate alkyl-di-imide (2) as already suggested³ for oxidations by hexacyanoferrate(III) and periodate. Such an alkyl-di-imide could be formed from the hydrazine either directly by two-electron oxidation¹ (as with hexacyanoferrate and periodate) or indirectly *via* an *N*-iodohydrazine (by analogy with the mechanism suggested⁶ for the oxidation of hydrazine itself by iodine).

The yield of alkane obtained from oxidations in non-aqueous media is seldom greater than 20%, the principal product usually being the corresponding alkyl iodide. This latter compound could be derived from either an

³ D. J. Cram and J. S. Bradshaw, *J. Amer. Chem. Soc.*, 1963, **85**, 1108.

⁴ D. M. Brown and G. H. Jones, *J. Chem. Soc. (C)*, 1967, 252.

⁵ Preliminary report, D. M. Brown and G. H. Jones, *Chem. Comm.*, 1965, 561.

⁶ H. Rottendorf and S. Sternhell, *Austral. J. Chem.*, 1963, **16**, 651.

intermediate diazonium iodide (5) formed in turn by two-electron oxidation of the alkyl-di-imide (2), or an intermediate *N*-iodo-compound (4) or (6) formed by

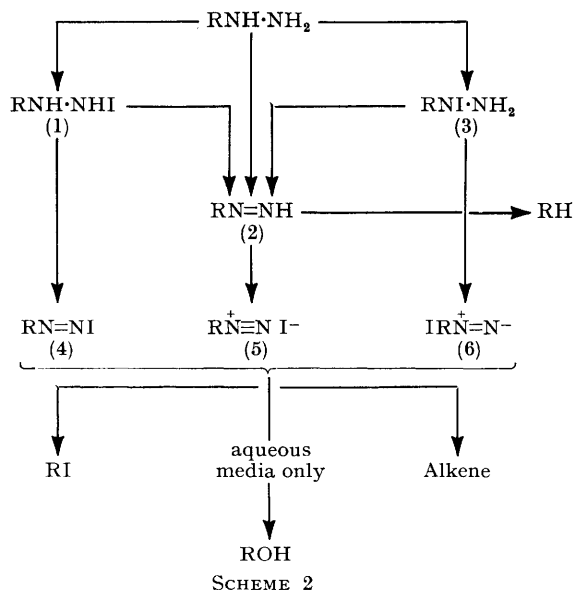
an ether as solvent. Even under such conditions, however, the actual yield of iodide is markedly dependent upon the structure of the alkylhydrazine. Thus, the

TABLE 1
Oxidation of alkylhydrazines by iodine

Expt. no.	R in RNH·NH ₂	Base	Solvent	Procedure ^a	Products (%)			
					RH	Alkene	RI	ROH
(i) Non-aqueous media								
1	Heptyl	Et ₃ N	CHCl ₃	A	<i>b</i>		52	
2	Heptyl		Et ₂ O	A	15		80	
3	Benzyl		Et ₂ O	A	12		85	
4	Phenethyl		Et ₂ O	A	8	1	88	
5	1-Methylpentyl		Dioxan	A	19		81	
6	Cyclohexyl		CHCl ₃	A	<i>b</i>	<i>b</i>	47	
7	Cyclohexyl		Et ₂ O	A	<i>b</i>	<i>b</i>	72	
8	Cyclopentyl		Et ₂ O	A	<1	29	71	
9	<i>t</i> -Butyl		Et ₂ O	B	<i>b</i>	<i>b</i>	6	
10	1,1-Dimethylpentyl		PhOMe	B	34	61		
11	1-Adamantyl		Et ₂ O	B	<1		95	
(ii) Aqueous media								
12	Heptyl	KOH	H ₂ O	C	25		13	18
13	Heptyl	CO ₃ ²⁻ -HCO ₃ ⁻		C	14		19	26
14	Heptyl	CO ₃ ²⁻ -HCO ₃ ⁻		D	11		67	21
15	Phenylethyl	Et ₃ N		C	10	2	75	10
16	(1-Methylpentyl)	CO ₃ ²⁻ -HCO ₃ ⁻		C	17		65	18
17	Cyclohexyl	CO ₃ ²⁻ -HCO ₃ ⁻		C	<i>b</i>	<i>b</i>	45	12
18	Cyclohexyl	CO ₃ ²⁻ -HCO ₃ ⁻		D	<i>b</i>	<i>b</i>	57	15
19	<i>t</i> -Butyl	KOH		C	<i>b</i>	<i>b</i>		63
20	<i>t</i> -Butyl	CO ₃ ²⁻ -HCO ₃ ⁻		C	<i>b</i>	<i>b</i>		80
21	1,1-Dimethylpentyl	KOH		C	41	14		45
22	1,1-Dimethylpentyl	CO ₃ ²⁻ -HCO ₃ ⁻		C	23	42		35
23	1,1-Dimethylpentyl	CO ₃ ²⁻ -HCO ₃ ⁻		D	26	41		33
24	1-Adamantyl	KOH		C	58		<1	42
25	1-Adamantyl	CO ₃ ²⁻ -HCO ₃ ⁻		C	26		5	69
26	1-Adamantyl	CO ₃ ²⁻ -HCO ₃ ⁻		D	36		9	55

^a See Experimental section. ^b Compound presumably formed but yield not determined since not separated from solvent under g.l.c. conditions.

two-electron oxidation of the *N*-iodohydrazines (1) and (3) postulated above as intermediates in the formation of the alkyl-di-imide (2).



The yield of alkyl iodide from a given hydrazine is usually greatest when oxidation is carried out with triethylamine as base and either chloroform or, better,

yields are generally high (>70%) when the hydrazino-group is attached to a primary or secondary carbon atom (experiments 2–5, 7, and 8 in Table 1), but negligible when it is attached to a tertiary non-bridgehead carbon atom, the principal products in these latter cases being alkenes (experiments 9 and 10). Such alkenes could well be formed from any of the possible intermediates (4)–(6) shown in Scheme 2 *via* either an *E*₁ or an *E*₂ mechanism. In either event, however, we would expect a prohibitively high activation energy when the hydrazino-group of the alkylhydrazine is attached to a tertiary bridgehead carbon atom, and in this regard it is of particular interest that 1-adamantylhydrazine affords an almost quantitative yield of the corresponding iodide on oxidation in ether (experiment 11).

(ii) *Oxidations in Aqueous Media*.—Except for the case of 1-adamantylhydrazine, the yield of alkane obtained from a given alkylhydrazine on oxidation in aqueous media is similar to that obtained by using non-aqueous media, provided that a weak base (CO₃²⁻-HCO₃⁻ or Et₃N) is used. If a strong base (KOH) is used, however, the yield of alkane is significantly greater (compare experiments 2, 12, and 13; also compare experiments 10, 21, and 22). In the case of the adamantylhydrazine, the yield of alkane is considerably greater when an aqueous medium is used even when the

base used is weak (compare experiments 11, 24, and 25). These facts are consistent with the involvement of a carbanion mechanism in alkane formation, as suggested earlier.

As with oxidations in non-aqueous media, oxidations under aqueous conditions only afford alkyl iodides when the hydrazino-group of the alkylhydrazine is attached to a primary, secondary, or tertiary bridgehead carbon atom. In each of these situations, the yield of alkyl iodide is greatest when a weak base is used, but even then the yield is lower than that under non-aqueous conditions, the effect being most marked in the case of 1-adamantylhydrazine (compare experiments 2, 12, and 13; 4 and 15; 5 and 16; 7 and 17; and 11, 24, and 25). The reduction in yield of alkyl iodide, which is largely due to simultaneous formation of the corresponding alcohol, can be offset by adding an excess of potassium iodide to the reaction mixture prior to addition of the iodine, especially when the hydrazino-group is attached to a primary carbon atom (compare experiments 13 and 14; 17 and 18; and 25 and 26).

Alcohol formation is also an important pathway when the hydrazino-group is attached to a tertiary non-bridgehead carbon atom, but in such cases the alcohols are formed largely at the expense of alkenes (compare experiments 10, 21, and 22).

(iii) *Stereochemistry and Mechanism of Alkyl Iodide Formation*.—As previously reported,⁴ oxidation of 3-deoxy-3-hydrazino-1,2,5,6-di-*O*-isopropylidene-*D*-allofuranose by iodine in chloroform affords the corresponding 3-deoxy-3-iodo-*D*-glucofuranose in high yield. In this case, therefore, alkyl iodide formation proceeds with inversion of configuration. However, since the hydrazino-sugar used in this particular work contained structural features which might themselves control the stereochemistry of oxidation, it was decided to investigate the stereochemistry of the oxidation of (1-methylheptyl)hydrazine, in which steric effects were expected to be minimal.

Two possible routes to the required optically active hydrazine were investigated. In the first, optically active octan-2-ol was converted into the toluene-*p*-sulphonate ester, which was then treated with hydrazine hydrate in ethanol or, better, with anhydrous hydrazine. In the second, the optically active alcohol was converted into the corresponding iodide, which was then treated with anhydrous hydrazine. This second method proved to be the better: although the overall yields were essentially the same, the specific rotation of the iodide was greater when the second method was used.

The second method of preparation also offers another advantage. Thus, if we assume that the reaction between 2-iodo-octane and hydrazine proceeds with complete inversion of configuration, then the stereochemical course of the oxidation of the resulting alkylhydrazine to the corresponding iodide may be determined by simple comparison of the specific rotation of this latter iodide with that of the iodide from which the alkylhydrazine was obtained initially. Typical results

obtained by this approach are given in Table 2. They show that alkyl iodide formation proceeds with predominant retention of configuration, thereby suggesting that a reaction pathway which involves an *S_N1*-type rearrangement of one or other of the postulated intermediate iodo-compounds [(4) and (6)] may well be important.

TABLE 2

Oxidation of optically active (1-methylheptyl)hydrazine by iodine

Expt. no.	Base	Solvent	Procedure ^a	Iodide obtained	
				[α] _D (°)	% Retention ^b
1	Et ₃ N	Et ₂ O	A	+21	73
2	Et ₃ N	CHCl ₃ ^c	A	+21	73
3	CO ₃ ²⁻ -HCO ₃ ⁻	H ₂ O	C	+31	84
4	CO ₃ ²⁻ -HCO ₃ ⁻	H ₂ O	D	+12.5	64
5	KOH	H ₂ O	C	+30	83
6	KOH	H ₂ O	D	+12.5	64

^a See Experimental section. ^b The hydrazine used was obtained from 2-iodo-octane having [α]_D -45°. For method of calculation of % retention see text. ^c Ethanol-free.

Competing with such a pathway, however, is another which results in inversion of configuration and which, in the case of oxidations in aqueous media, assumes greater importance when iodide ion is added to the reaction mixture prior to addition of the oxidant (compare experiments 3 and 4; and 5 and 6 in Table 2). This second pathway may therefore involve an *S_N2*-type reaction of one or other of the same intermediate iodo-compounds [(4) and (6)]. Alternatively, an *S_N1*-type reaction may be involved, but this seems less likely to be true for alkylhydrazines in general since, as already noted, the increase in yield of alkyl iodide resulting from addition of iodide ion prior to addition of the oxidant is greatest when the hydrazino-group of the alkylhydrazine is attached to a primary atom.

The stereochemical course of alkyl iodide formation observed in the present work in the case of (1-methylheptyl)hydrazine is clearly different from that observed previously in the sugar series,⁴ conceivably owing to steric effects peculiar to the hydrazino-sugar. Further studies of such effects are in progress.

(iv) *Stereochemistry and Mechanism of Alcohol Formation*.—In the course of the above studies of the oxidation of (1-methylheptyl)hydrazine to the corresponding iodide, it was necessary to separate the iodide from the corresponding alcohol. Examination of this alcohol showed that, in all cases, it was optically inactive. It is therefore possible that although alkyl iodide formation is not likely to proceed *via* an intermediate carbocation (see above), such ions may well be intermediates in alcohol formation.

EXPERIMENTAL

I.r. spectra were recorded with a Perkin-Elmer 257, a Perkin-Elmer Infracord, or a Unicam SP 200 spectrometer. ¹H N.m.r. spectra were recorded with either a Perkin-Elmer 60 or a Varian A56/60 spectrometer (internal tetramethyl-

silane standard). Mass spectra were recorded with either an A.E.I. MS-9 or a Hitachi-Perkin-Elmer RMU-6E spectrometer.

G.l.c. was carried out by using the following instruments: (1) F and M. 609, nitrogen carrier gas; (2) F and M 720, hydrogen carrier gas; (3) Varian Aerograph A-700, helium carrier gas. The following columns were used: PPE (10% polyphenyl ether on 60–80 mesh Chromosorb W, 6 ft \times 1/4 in), SR (20% SE-30 silicone rubber on 60–80 mesh Chromosorb W, 2 ft \times 1/4 in), CW-1 (12% Carbowax 4000 on 100–120 mesh GasChrom CLP, 10 ft \times 1/4 in), CW-2 (8% Carbowax 4000 on 100–120 mesh GasChrom CLP, 6 ft \times 1/4 in), APL (5% Apiezon L on GasChrom CLH, 5 ft 6 in \times 1/4 in), and NU (11% Nujol on 100–120 GasChrom CLP, 10 ft \times 1/4 in).

Optical rotations were measured by using a Rudolph 500 polarimeter with a sodium lamp as light source: compounds were examined either as neat liquids or as *ca.* 2% solutions in the solvent indicated.

Preparation of Hydrazines.—(a) *Heptylhydrazine.* Heptyl iodide (15 ml) and hydrazine hydrate (25 ml) were refluxed, under nitrogen, for 30 min in ethanol (35 ml). The cooled mixture was treated with aqueous potassium hydroxide (10%; 50 ml) and then extracted with ether (3 \times 50 ml). The combined extracts were dried (Na_2SO_4) and evaporated, and the residue distilled to give the required hydrazine (6 g, 51%), b.p. 58° at 0.5 mmHg. On treatment with ethanolic anhydrous oxalic acid it afforded an *oxalate salt*, m.p. 168° (from water) (Found: C, 49.0; H, 9.2. $\text{C}_9\text{H}_{20}\text{N}_2\text{O}_4$ requires C, 49.0; H, 9.1%).

(b) *Benzylhydrazine.* The free hydrazine (38%; b.p. 82° at 0.5 mmHg) was obtained from benzyl chloride by the method described above. The *oxalate salt* had m.p. 192° (from water) (lit.,⁷ 182–183°) (Found: C, 50.9; H, 5.8; N, 13.6. Calc. for $\text{C}_9\text{H}_{12}\text{N}_2\text{O}_4$: C, 51.0; H, 5.7; N, 13.4%).

(c) *Phenethylhydrazine.* The free hydrazine was obtained from the commercially available sulphate salt.

(d) *(1-Methylpentyl)hydrazine.* The free hydrazine was obtained from the corresponding iodide by the method described for heptylhydrazine. The *oxalate salt* had m.p. 140–141° (from aqueous ethanol) (Found: C, 46.3; H, 8.6; N, 13.8. $\text{C}_8\text{H}_{18}\text{N}_2\text{O}_4$ requires C, 46.6; H, 8.8; N, 13.6%).

(e) *Cyclohexylhydrazine.* This compound was obtained by catalytic reduction of cyclohexanone hydrazone.⁸ The *oxalate salt* had m.p. 210° (from water) (lit.,⁹ 196–197°) (Found: C, 47.2; H, 7.5; N, 13.4. Calc. for $\text{C}_8\text{H}_{16}\text{N}_2\text{O}_4$: C, 47.0; H, 7.9; N, 13.7%).

(f) *Cyclopentylhydrazine.* The free hydrazine, b.p. 65–70° at 11 mmHg (lit.,¹⁰ b.p. 60–65° at 11 mmHg), was obtained similarly from cyclopentanone hydrazone. The *oxalate salt* had m.p. 163° (from aqueous ethanol) (Found: C, 44.1; H, 7.1. $\text{C}_7\text{H}_{14}\text{N}_2\text{O}_4$ requires C, 44.4; H, 6.9%).

(g) *t-Butylhydrazine.* The hydrochloride was prepared in 2% yield by reaction of t-butyl chloride with hydrazine as described by Westphal.¹¹ On treatment with aqueous sodium hydroxide it afforded the free hydrazine, which was extracted with ether. The extract was added immediately to saturated ethanolic oxalic acid; the *oxalate salt* had m.p. 176–177° (from aqueous ethanol) (Found: C, 40.2;

H, 7.8; N, 15.7. $\text{C}_6\text{H}_{14}\text{N}_2\text{O}_4$ requires C, 40.4; H, 7.9; N, 15.7%).

(h) *(1,1-Dimethylpentyl)hydrazine.* The *hydrochloride* was prepared in 2% yield by essentially the same method as that for t-butylhydrazine; m.p. 94–95° (Found: C, 50.2; H, 11.5; N, 16.9. $\text{C}_7\text{H}_{16}\text{ClN}_2$ requires C, 50.4; H, 11.5; N, 16.8%).

(i) *(1-Adamantyl)hydrazine.* A solution of ethyl bromoacetate (5.5 g, 0.033 mol) in benzene (10 ml) was added slowly to a hot solution of 1-adamantylamine (10 g, 0.066 mol) in benzene (50 ml). The mixture was refluxed for 2 h and cooled, and the amine hydrobromide filtered off. The filtrate was evaporated under reduced pressure and the residual N-substituted glycine ester was then heated under reflux with sodium hydroxide (5 g) in water (50 ml) for 30 min. The cooled mixture was acidified to pH 2 with conc. hydrochloric acid and the precipitated N-(1-adamantyl)glycine was collected; yield 5.5 g (80%), decomp. 290° (lit.,¹² decomp. 300°). The glycine was converted successively into N-nitroso-N-(1-adamantyl)glycine (m.p. 164°; lit.,¹² 158°), 3-(1-adamantyl)sydnone (m.p. 215–215.5°; lit.,¹² 215–217°), and (1-adamantyl)hydrazine by the method of Daeniker.¹² The hydrazine gave an *oxalate salt*, m.p. 210–211° (decomp.) (Found: C, 56.0; H, 8.1; N, 10.7. $\text{C}_{12}\text{H}_{20}\text{N}_2\text{O}_4$ requires C, 56.2; H, 7.9; N, 10.9%).

Oxidation of Hydrazines; General Methods.—(i) *Method A.* The hydrazine (1 mmol) was dissolved in the appropriate solvent (5–10 ml) and the appropriate base (3 mmol) was added. The flask was flushed with nitrogen and the mixture was stirred while solid iodine was added until present in slight excess as indicated by cessation of effervescence; 2 mmol of iodine was usually required. The mixture was made up to a standard volume (25 ml) and duplicate analyses were then performed by g.l.c. (columns, conditions, and retention times are shown in Table 3). Yields of the various products (Table 1) were determined by comparison of peak areas with those obtained from standard solutions of authentic samples (either available commercially or prepared by standard methods).

(ii) *Method B.* The procedure of method A was used except that the *oxalate salt* of the hydrazine was used.

(iii) *Method C.* The *oxalate salt* (1 mmol) of the hydrazine was dissolved in boiling water (10 ml) and the solution was cooled under nitrogen with continuous stirring in an ice-bath. The appropriate base (5 mmol) was then added, followed by solid iodine (*ca.* 2 mmol) until present in slight excess as indicated by cessation of effervescence. The brown colour of the solution was discharged by addition of a small amount of sodium thiosulphate and the products were extracted with ether (10 ml; then 2 \times 5 ml). The combined extracts were made up to a standard volume (25 ml) and analysed as in method A.

(iv) *Method D.* The procedure of method C was used except that potassium iodide (5 mmol) was added to the mixture before the iodine.

Stereochemical Studies.—*Preparation of (1-methylheptyl)hydrazine.* By the method of Coulson *et al.*,¹³ octan-2-ol was converted into the corresponding dialkyl hydrogen

⁷ R. Huisgen, F. Jakob, W. Siegel, and A. Cadus, *Annalen*, **1954**, **590**, 1.

⁸ J. Druey, P. Schmidt, and K. Eichenberger, Ger. Pat. 1,089,388 (*Chem. Abs.*, 1962, **57**, 4681).

⁹ E. Schmitz and R. Ohme, *Chem. Ber.*, 1961, **94**, 2166.

¹⁰ J. Druey, P. Schmidt, K. Eichenberger, and M. Wilhelm, Ger. Pat. 1,082,258 (*Chem. Abs.*, 1961, **55**, P 23384b).

¹¹ O. Westphal, *Ber.*, 1941, **74B**, 759.

¹² H. U. Daeniker, *Helv. Chim. Acta*, 1967, **50**, 2008.

¹³ E. J. Coulson, W. Gerrard, and H. R. Hudson, *J. Chem. Soc.*, 1965, 2364.

phosphite, b.p. 137° at 0.5 mmHg (lit.,¹³ 135–144° at 2 mmHg), which was then converted into 2-iodo-octane, b.p. 54° at 0.3 mmHg (lit.,¹³ 41° at 0.5 mmHg); overall yield 16%. The yield was higher (54%) when the intermediate phosphite was not isolated. The iodide (8.8 g) and anhydrous hydrazine (9 ml) were heated under reflux in ethanol (30 ml) for 20 min. The crude free hydrazine was isolated in the usual way (see earlier) and distilled;

132 °C). The indicated g.l.c. conditions were sufficiently mild to enable authentic optically active 2-iodo-octane to pass through the column with minimal loss of optical activity. The degree of racemisation of the authentic sample under these conditions was used as a basis for correcting the observed optical activity of the 2-iodo-octane isolated from oxidation experiments. Such corrected rotations are shown in Table 2.

TABLE 3
G.l.c. analysis of oxidation products

Hydrazine	Column (instrument ^a)	Temp. (°C)	Flow rate (ml min ⁻¹)	Retention times (min)			
				RH	>C=C<	ROH	RI
Heptyl	PPE (2)	40	75	2.8	3.0		
		135	60			2.6	5.6
Phenethyl	SR (2)	90	110	0.9	1.1	4.7	14.2
1-Methylpentyl	CW-1 (1)	<i>b</i>	30	1.3	1.6	5.2	11.4
Cyclohexyl	CW-1 (1)	110	40	1.2	1.5	7.9	16.4
Cyclopentyl	NU (1)	50	45	4.8	3.8		
	CW-2 (1)	110	40				6.4
t-Butyl	CW-1 (1)	70	40			4.4	5.0
1,1-Dimethylpentyl	CW-1 (1)	100	40	1.1	1.3	8.4	12.0
1-Adamantyl	APL (1)	160	50	1.7		4.0	11.5

^a See Experimental section. ^b Column temp. 55 °C for 2 min, then 110 °C.

b.p. 65° at 0.5 mmHg; yield 72%. The oxalate salt had m.p. 134° (lit.,¹⁴ 133–134°).

When optically active octan-2-ol {[α]_D + 6.65° (neat liquid)} was used as starting material, the iodide had [α]_D – 45° (in chloroform), the free hydrazine [α]_D – 10.6° (in ethanol), and the oxalate salt [α]_D – 12.0° (in ethanol).

Oxidation of the hydrazine. Oxidations of optically active (1-methylheptyl)hydrazine and its oxalate salt were carried out by the procedures described above for the other oxidations, except that in method A the reaction mixture was shaken with aqueous sodium thiosulphate and then with 3M-hydrochloric acid before separation of the products by preparative g.l.c. (F and M 720; 60 ml min⁻¹ hydrogen; 2 ft × 1/4 in QF1 column at 90 °C; injection port temp.

Effect of oxidation conditions on products. Separate experiments established that optically active 2-iodo-octane suffered no significant loss of activity under the conditions used in any of the oxidation experiments.

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¹⁴ E. Michel-Ber, F. Dvolaitzky, and J. Guilbert, Brit. Pat. 899,385 (*Chem. Abs.*, 1963, **58**, P 1346f).