

TABLE VII
Crystallization of Lipid Glycerides of *V. anthelmintica* Seed Oil at
-60C for Removal of Unsaponifiable Material

	Quantity	Oxirane oxygen	FFA as epoxy-oleic	Iodine value (Wijs)	Unsaponifiable material
	g	%	%		%
Oil.....	1698	3.85	0.86	104.1	6.67
Precipitate.....	1511	4.39	0.52	87.8	1.20
Filtrate.....	188	0.29	2.4	190.0	53.0

of flaking dry seed at room temp, followed by rapid handling in the extraction steps, was not satisfactory from the standpoint of producing an oil with a low free fatty acid content. Even when the flaked seed was allowed to fall directly from the rollers of the mill into the solvent this was not successful in holding down lipolysis.

The process of autoclaving the seed to inhibit lipolysis prior to flaking also was a successful procedure in oil production as illustrated by the fractional extraction data obtained. The slight variation in the free fatty acid content and oxirane oxygen composition of the 10 extracts may have been due to selective extraction of the epoxy fatty components. When the seed was autoclaved no haste was necessary in carrying out extraction operations.

In situations where trivernolin was the desired end-product (Tables V and VI) the rapid technique offered several advantages over the autoclaved seed treatment: the yield of good-quality trivernolin was

higher; less coloring material was extracted; and it was not necessary to concn the solvent before crystallization in order to obtain the product in good yield. The solubility of trivernolin has previously been reported (4) as ca. 5 g/gal at -20C.

No plastic formulation data are available on the evaluation of *Vernonia* oil where the major portion of the unsaponifiable material has been removed. As this process appreciably elevated the oxirane oxygen percentage, reduced the I.V., and removed color and odor, it may have resulted in an improved product for the purpose stated.

Since the quantity of oil extracted by Waring Blendor technique, 27.9%, is in close agreement with the analytical figure of 27.2% obtained by tedious, exhaustive Soxhlet extraction (1) the former method is now recommended for estimation of the quantity of oil in *Vernonia* seed. Additional experiments have confirmed the utility of this procedure.

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Physical Properties of Fatty Acid Methyl Esters. II. Refractive Index and Molar Refraction.

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Abstract

The refractive indices of methyl oleate, linoleate, linolenate, erucate, and the saturated fatty acid methyl esters from acetate to nonadecanoate have been measured at 20C and 40C for the N_D , H_α , H_β , and H_γ lines. The values for the saturated series have been correlated with the Smittenberg relation. Molar refractions have been computed and checked for additivity. The limiting refractive indices obtained from the Smittenberg relation are compared to those obtained from the molar refraction.

Introduction

THE REFRACTIVE INDEX, which may be determined with relative ease, is an important aid for identification and for a test of purity of organic compounds. For constitutive studies, however, it is in the molar refraction, which is directly related to structural and physical properties of the molecule, that the value of the refractive index is most apparent. For our investigations on the relationships between various physical and chemical properties of fatty oils and their derivatives (13,37) these two physical properties are, therefore, eminently suited to occupy a central place in our considerations. Pure model com-

pounds, such as the fatty acid methyl esters, may be used as basis for obtaining the accurate data requisite to the computation of reliable molar refractivity increments. There is already an abundance of data available on n_D^{20} , the refractive index of the N_D line at 20C, of fatty acid methyl esters. In 1954 Hammond and Lundberg published a paper on the molar refraction, molar volume, and refractive index of fatty acid esters and related compounds in the liquid state (15), which presented a comprehensive review of the subject as of that date. A noticeable drawback was that the cited data, which have been obtained from several sources, vary widely for some compounds. An additional deficiency is the scarcity of data at wavelengths and temp other than n_D^{20} .

For these reasons we have prepared some fatty acid methyl esters in high purity and measured the refractive index for the N_D line ($\lambda = 5892.6 \text{ \AA}$) and for the red (H_α , $\lambda = 6562.8 \text{ \AA}$), green (H_β , $\lambda = 4861.3 \text{ \AA}$), and violet (H_γ , $\lambda = 4340 \text{ \AA}$) hydrogen lines at 20C and 40C.

The investigated compounds were the saturated fatty acid methyl esters from acetate to nonadecanoate, methyl oleate, linoleate, linolenate, and erucate. The preparation, purity, and the density of these compounds have already been described in a previous communication (13).

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TABLE I
 Refractive Index of Saturated Fatty Acid Methyl Esters, 20C

Compound	Measured values				Literature values				Ref	
	H _α	N _{AD}	H _β	H _γ	H _α	N _{AD}	H _β	H _γ		
Acetate.....	1.35966	1.36139	1.36573	1.36915	1.36015	1.36193	1.36629	1.36930	(44)	
					1.35963	1.36133	1.36572	1.36840	(40)	
					1.3600	1.3615	1.3661		(16)	
					1.3606		1.3667	1.3693	(9)	
Propionate.....	1.37499	1.37685	1.38130	1.38482		1.3614			(14)	
						1.3619			(17)	
						1.3615			(11)	
						1.36143			(24)	
					1.37516	1.37750	1.38132	1.38482	(34)	
					1.37494	1.37683	1.38130	1.38500	(41)	
					1.37306	1.37683	1.38130	1.38437	(44)	
Butyrate.....	1.38562	1.38754	1.39226	1.39605		1.37643			(48)	
						1.37724			(23)	
					1.38357	1.38729	1.39193	1.39528	(44)	
Valerate.....	1.39546	1.39742	1.40227	1.40614	1.3854	1.3878	1.3919	1.3954	(34)	
					1.38693		1.39359	1.39742	(21)	
					1.39493	1.39690	1.40167	1.40505	(44)	
Caproate.....	1.40330	1.40535	1.41030	1.41429		1.39743			(1)	
					1.40300	1.40494	1.40996	1.41352	(44)	
Oenanthat.....	1.40970	1.41178	1.41681	1.42084	1.40298		1.40990		(39)	
						1.40536			(1)	
						1.40528			(31)	
						1.4052			(3)	
Caprylate.....	1.41504	1.41716	1.42227	1.42638	1.40946	1.41152	1.41654	1.42010	(44)	
					1.40925		1.41649		(39)	
						1.4113			(29)	
						1.41180			(1)	
Pelargonate.....	1.41958	1.42173	1.42690	1.43107		1.41696	1.42203	1.42574	(44)	
					1.41486				(3)	
						1.4170			(1)	
Caprate.....	1.42350	1.42569	1.43093	1.43519		1.41715			(31)	
						1.41708			(2)	
					1.4195	1.4216	1.4269		(1)	
Undecanoate.....	1.42693	1.42910	1.43444	1.43875		1.42179			(44)	
						1.42343	1.42556	1.43078	1.43456	(26)
						1.4237	1.4259	1.4310		(3)
						1.4256	1.42539			(25)
Laurate.....	1.42991	1.43209	1.43750	1.44185		1.42573			(1)	
					1.42971	1.42908			(1)	
Tridecanoate.....	1.43247	1.43470	1.44012	1.44451		1.43188	1.43720	1.44114	(44)	
						1.43207			(1)	
						1.43171			(25)	
						1.4318			(3)	
						1.4320			(22)	
						1.43470			(1)	
						1.43702			(1)	
Myristate.....	1.43480	1.43701	1.44248	1.44683		1.43683			(25)	
						1.4370			(3)	
						1.4368			(22)	
						1.4369			(46)	
Pentadecanoate.....	1.43690	1.43914	1.44464	1.44902		1.4390			(32)	
						1.43909			(1)	

 TABLE II
 Refractive Index of Saturated Fatty Acid Methyl Esters; 40C

Compound	Measured values				Literature values				Ref
	H _α	N _{AD}	H _β	H _γ	H _α	N _{AD}	H _β	H _γ	
Acetate.....	1.34943	1.35116	1.35532	1.35859		1.35193			(40)
Propionate.....	1.36499	1.36681	1.37119	1.37467	1.36494	1.36680	1.37130		(41)
Butyrate.....	1.37599	1.37786	1.38239	1.38597					
Valerate.....	1.38621	1.38814	1.39284	1.39663					
Caproate.....	1.39432	1.39631	1.40115	1.40504					
Oenanthat.....	1.40099	1.40301	1.40796	1.41189					
Caprylate.....	1.40650	1.40857	1.41353	1.41762					
Pelargonate.....	1.41128	1.41336	1.41842	1.42256					
Caprate.....	1.41531	1.41741	1.42256	1.42669		1.4174			(25)
Undecanoate.....	1.41881	1.42095	1.42616	1.43034					
Laurate.....	1.42186	1.42401	1.42924	1.43346		1.4239			(25)
						1.4238			(27)
Tridecanoate.....	1.42447	1.42665	1.43192	1.43615					
Myristate.....	1.42684	1.42905	1.43436	1.43867		1.4290			(25)
Pentadecanoate.....	1.42902	1.43122	1.43659	1.44092					
Palmitate.....	1.43094	1.43318	1.43856	1.44286		1.4330			(25)
						1.43314			(7)
Heptadecanoate.....	1.43269	1.43492	1.44033	1.44460					
Stearate.....	1.43426	1.43651	1.44195	1.44632		1.43649			(7)
						1.4365			(25)
						1.4370			(28)
Nonadecanoate.....	1.43570	1.43796	1.44342	1.44786					

 TABLE III
 Refractive Index of Some Unsaturated Fatty Acid Methyl Esters

Compound	Measured values								Literature		Ref
	20C				40C				20C	40C	
	H _α	N _{AD}	H _β	H _γ	H _α	N _{AD}	H _β	H _γ	N _{AD}		
Oleate.....	1.44962	1.45214	1.45834	1.46339	1.44203	1.44450	1.45052	1.45549	1.45195	1.4443	(25)
									1.4522		(45)
									1.45214	1.44460	(7)
										1.4445	(28)
Linoleate.....	1.45871	1.46150	1.46841	1.47404	1.45109	1.45385	1.46060	1.46615	1.4521		(19)
									1.4523		(20)
									1.4612		(30)
									1.4613		(36, 43)
Linolenate.....	1.46825 ^a	1.47133	1.47896	1.48530	1.46074	1.46381	1.47130	1.47752	1.4616		(25)
									1.4709		(43)
Erucate.....	1.45332	1.45583	1.46200	1.46709	1.44581	1.44828	1.45435	1.45940	1.4711		(25)
									1.4558		(42)
									1.45558	1.44812	(7)

^a Contains 27.5% *trans* double bonds.

Experimental

Measurements of the refractive index were carried out on a Pulfrich refractometer. Distilled water was used as the calibrating agent and the values presented by Tilton and Taylor (38) accepted as standards. Purified benzene was used as a secondary check for which the values of Barbaudy (4) and of Campbell and Miller (6) were applied. Temp readings were carried out with a calibrated thermometer and the obtained values corrected for the protruding mercury thread. In general, three readings were made of each point. More readings were usually necessary for the H_γ line. Due to the very weak character of the band, readings could not be effected with a high degree of precision, and in comparison to the other measurements the accuracy of the H_γ values is, therefore, much lower.

The obtained values show in Tables I, II, and III, and compared with some values from the literature. Large differences are sometimes noted. In many cases, however, it is generally possible to doubt the purity of these compounds. Values which deviate significantly from the average values have been deleted.

A good concurrence is generally observed with data with a high degree of reliability, e.g., with the values of Adriaanse (1), whose compounds are purer than 99.6% as derived from melting graphs.

The Smittenberg Relations. The relation was first proposed by Smittenberg and Mulder (35)

$$y = y_\infty + A/(B + n) \quad [1]$$

and can be applied to the eight series obtained for the saturated fatty acid methyl esters. In this relation:

y = physical constant, in this particular case the refractive index,

y_∞ = the limiting value of this physical constant,

n = the number of C-atoms in the fatty acid chain, A and B are constants.

y_∞ , A, and B have been calculated assuming two alternatives:

- 1) All measured terms are taken into consideration.
- 2) The lower members are neglected and only those terms from methyl caproate and higher are considered. For the Na_D -series at 20C the values of A and B have also been calculated assuming $n_{D,\infty}^{20}$ to be 1.4752, the limiting value of the hydrocarbons (35). The obtained values are presented in Table IV.

Assuming the Smittenberg relation to be essentially correct, an estimate of the contribution of error from the measurements and from the impurities in the samples may be obtained from the variance estimate

$$\sigma^2 = \delta^2/(m - 3) \quad [2]$$

σ = standard deviation,

δ = difference between measured and computed values,

m = number of terms considered.

The data obtained from this relation can be applied to an analysis of variance for the resolution of errors.

To compare the relative merits of the obtained numerical values it will be necessary to apply the Variance Ratio Test to any two alternative type relations. Reference is made to the previous communication where the density has been subjected to

TABLE IV
Numerical Values for the Smittenberg Relation for Saturated Fatty Acid Methyl Esters

Temp	n	Line	y_∞	A	B	σ
20C	≅2	H_α	1.47701	-0.78793	4.6948	0.000278
		Na_D	1.47941	-0.79053	4.6783	0.000286
		H_β	1.48572	-0.80742	4.7089	0.000269
40C	≅2	H_γ	1.49058	-0.81600	4.6970	0.000266
		H_α	1.46910	-0.78185	4.4874	0.000363
		Na_D	1.47163	-0.78852	4.4999	0.000363
20C	≅6	H_β	1.47762	-0.80114	4.5052	0.000370
		H_γ	1.48237	-0.80937	4.4923	0.000385
		H_α	1.47472	-0.72332	4.1238	0.000025
40C	≅6	Na_D	1.47706	-0.72436	4.0969	0.000021
		H_β	1.48348	-0.74438	4.1662	0.000034
		H_γ	1.48799	-0.74259	4.0645	0.000060
20C	≅5	H_α	1.46721	-0.72159	3.8979	0.000030
		Na_D	1.46977	-0.72922	3.9252	0.000025
		H_β	1.47579	-0.74318	3.9550	0.000029
40C	≅5	H_γ	1.48057	-0.75210	3.9560	0.000042
		Na_D	1.4752	-0.67417	3.6358	0.000081

this test. It will be also apparent that for the refractive index significant deviations are observed if the lower members of the homologous series are included in the calculations. Geelen (12) described the relation of Smittenberg as a transformation of the function

$$y = y_\infty + A/n + B/n^2 + C/n^3 + D/n^4 + \quad [3]$$

in which the fourth and consecutive terms are neglected. It will be apparent that for the higher homologs this assumption is generally quite permissible, but that for the first members of the series appreciable deviations may occur. This phenomenon has been observed in the densities and in other physical constants under investigation.

The Molar Refraction. The molar refraction according to Lorentz-Lorenz,

$$R_m = (n^2 - 1)M/(n^2 + 2)d \quad [4]$$

has been computed for the investigated compounds. In this equation n is the refractive index, M the molecular weight, and d the density. CH_2 increments have been computed as differences between consecutive members of the saturated series. Except for the lower members, where (as already noted by Eykman in 1895) appreciable deviations occur, this value is constant for the whole investigated range.

A closer look at the increments, however, indicates the existence of a slight trend of R_{CH_2} decreasing with increasing chain length. The inconsistency of R_{CH_2} , the CH_2 increment, has been noted before. Weber et al. (33) demonstrated that R_{CH_2} is not completely independent of the molecular weight. Although the noted deviations are still within the limits of experimental error it seems advisable that for precise determinations one should avail oneself of the more general relation

$$R_m = A + nR_{CH_2} f(n) \quad [5]$$

Assuming that CH_2 increment to be constant (which will only introduce a small error) use can be made of the relation

$$R_m = A + nR_{CH_2} \quad [6]$$

to correlate the values of the molar refraction.

From the observed data the eight regression equations have been computed. To obviate the effect of the primary terms only methyl caproate and the higher terms are taken into consideration. Numerical values for the regression equations show in Table V. The product moment correlation coefficient is practically 1 for all observed series. R_{CH_2} is, therefore, the statistical average of the computed CH_2 increments. Some comparative data are presented in Table VI. Due to the abundance of available values a rather random selection has been made.

TABLE V
Numerical Values in Regression Equations for Saturated
Fatty Acid Methyl Esters

Temp	Line	A	R _{CH₂}	σ
20C	H _α	8.228	4.6203	0.0040
	Na _D	8.267	4.6407	0.0046
	H _β	8.345	4.6920	0.0040
	H _γ	8.413	4.7324	0.0070
	40C	H _α	8.278	4.6262
	Na _D	8.311	4.6473	0.0100
	H _β	8.398	4.6977	0.0097
	H _γ	8.468	4.7380	0.0088

Assuming a linear behaviour in the investigated range the temperature dependence of the CH₂ increment may be assessed from

$$H_{\alpha} : R_{CH_2} = 4.614 + 0.0003 t \quad [7]$$

$$Na_D : R_{CH_2} = 4.634 + 0.00033 t \quad [8]$$

$$H_{\beta} : R_{CH_2} = 4.686 + 0.0003 t \quad [9]$$

$$H_{\gamma} : R_{CH_2} = 4.727 + 0.00028 t \quad [10]$$

where t is the temperature in degrees C.

Unsaturation. An estimate of the increment of the double bond, R_f, may be obtained from the molar refractions of the unsaturated compounds in our investigations and from methyl stearate and behenate. Molar refractions of these saturated esters can be obtained from the regression equations, numerical values for which are presented in Table V. R_f was computed as the difference between the molar refractions of stearate and oleate, oleate and linoleate, and behenate and erucate. Due to the high percentage of *trans* double bonds in methyl linolenate the difference between this compound and methyl linoleate may not be applied for computing R_f. This value is also significantly different in comparison to the other observed R_f values.

As already noted previously (13,15) it seems that the increment of the first double bond is not equal to the contribution of additional non-conjugated double bonds in the same chain.

The averaged values and some comparative data show in Table VII. Due to the small number and due to the low precision of these measurements no values are suggested for H_γ.

Limiting Value of the Refractive Index. The molar refraction of the members of a homologous series may be represented by the relation:

$$R_m = A + nR_{CH_2} \quad [11]$$

The mol wt may be represented by a relation identical to this, i.e.,

$$M = C + 14.026 n \quad [12]$$

The specific refraction may be obtained by combining these two relations, i.e.,

$$r_{sp} = R_m/M = (A + nR_{CH_2})/(C + 14.026 n) \quad [13]$$

From this relation the limiting value of the specific refraction is obtained as

$$\lim_{n \rightarrow \infty} r_{sp} = R_{CH_2}/14.026 = (n_{\infty}^2 - 1)/(n_{\infty}^2 + 2) d_x \quad [14]$$

TABLE IV
CH₂ Increments at 20C

Author	R _{CH₂}				Year	Ref
	H _α	Na _D	H _β	H _γ		
Eisenlohr.....	4.598	4.618	4.668	4.710	1910	(8)
Fajans and Knorr.....		4.64			1926	(10)
Wibaut and Langedijk.....		4.639			1940	(47)
Vogel.....	4.624	4.648	4.695	4.735	1946	(18,44)
Tels et al.....		4.643			1958	(37)
Adriaanse.....		4.641			1960	(1)

TABLE VII
Increment of the Double Bond at 20C

Author	H _α	Na _D	H _β	H _γ	Year	Ref
Eisenlohr.....	0.498	0.467	0.406	0.351	1910	(8)
Fajans & Knorr.....		0.47			1926	(10)
Vogel.....	0.507	0.481	0.413	0.360	1948	(44)
Hammond & Lundberg.....		0.30			1954	(15)
Tels et al.....		0.24			1958	(37)
This work.....	0.40	0.35	0.27			

The limiting values of the density of the saturated fatty acid methyl esters, i.e., 0.85407 at 20C and 0.84225 at 40C, have already been reported previously (13). Substitution of these values in equation 14 yields the limiting refractive index.

	20C	40C
H _α	1.47460	1.46764
Na _D	1.47704	1.47010
H _β	1.48323	1.47607
H _γ	1.48810	1.48085

Comparison of these values with the data obtained from the Smittenberg relation (Table IV) indicates a large degree of agreement although two different approaches have been applied. Especially the values at 20C demonstrate a remarkable concurrence, but the excellent fit is probably fortuitous.

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