Density Estimation for Fatty Acids and Vegetable Oils Based on Their Fatty Acid Composition

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The liquid density of fatty acids can be accurately estimated by the modified Rackett equation over a wide range of temperatures. The modified Rackett equation requires the critical properties and an empirical parameter, Z_{RA}, for each acid as the basis for computing density as a function of temperature. The liquid density of vegetable oils can be estimated by using mixture properties corresponding to the fatty acid composition and a correction for the triglyceride form. The density prediction is explicitly temperature-dependent.

KEY WORDS: **Density, fatty acid, process design, physical property estimation, vegetable oil.**

Increasingly, modern process design involves the use of computer-aided process design techniques. The physical property packages used in chemical process simulators typically rely on generalized equations for predicting properties as a function of temperature, pressure, etc. The Rackett equation is commonly used for liquid density estimation. This paper demonstrates the use of the Rackett equation for fatty acids and vegetable oil liquid density estimation.

Two criteria are important for estimating the density of fatty acids. First, all of the commonly encountered saturated and unsaturated fatty acids must be included in the density estimation scheme. Second, the estimation scheme must account for the temperature dependency of density. Both criteria are important for the design of vegetable oil processing facilities and are frequently missing from previous fatty acid density estimation methods.

There have been numerous attempts to estimate the liquid density of fatty acids based on a linear relationship between the length of the fatty acid chain and its molar volume. Garner and Ryder (1) observed a 16.8 cc/gmole increase in liquid molar volume for each -CH_2- added to a fatty acid chain for acids 8:0–12:0 at 50°C. Dorinson *et al.* (2) developed equations that included most saturated fatty acids at specified temperatures. For the fatty acids 2:0 through 9:0, the molar volumes, V_m, at 20°C were given by:

$$V_m = 16.89n + 23.62 \qquad [1]$$

The molar volumes for the fatty acids 4:0–18:0 at 80°C were given by:

$$V_m = 17.25n + 28.88 \qquad [2]$$

where n is the number of carbon atoms in the fatty acid chain.

Hammond and Lundberg (3) expanded upon Dorinson *et al's* (2) work by adding terms to account for unsaturation and temperature dependence. Their equation for saturated and monounsaturated fatty acids is:

$$V_m = 16.54C - 6.65D + 26.09 + (0.006C + 0.0085)(T - 20) \qquad [3]$$

Hammond and Lundberg's Equation 3 for polyunsaturated fatty acids is:

$$V_m = 16.54C - 6.87D + 26.09 + (0.006C + 0.0085)(T - 20) \qquad [4]$$

where C is the number of carbon atoms, D is the number of double bonds in the fatty acid chain and T is the temperature in °C.

Costello and Bowden (4) used the Verschaffelt equation (Equation 5) to correlate saturated fatty acid density data, where d_l is the liquid density and d_g is the gas density:

$$d_l - d_g = d_o \left(1 - \frac{T}{T_c}\right)^m \qquad [5]$$

The two parameters, d_o and m, were fitted by using experimental data over a temperature range for individual fatty acids. The resulting equations gave a good representation of the data (reported to be generally with 0.3%). However, it was not suggested how this approach might be extended to estimate parameters without experimental data, particularly for d_g.

Work has been done by Fisher (5) in correlating fatty acid density with the carbon chainlength for the saturated fatty acids 4:0–30:0. This correlation is of the simple linear form:

$$d = b + m\,t \qquad [6]$$

where d is the density in g/cc, t is the temperature in °C, b is the intercept and m is the slope of the linear least-squares fit of the data. This correlation was developed for the temperature range of 20–80°C (5) and, thus, is limited in applicability. Also, the fatty acids 10:0–30:0 all have melting points either within the range or higher, so there is some question about the suitability of Equation 6 for these materials. More recent work by Fisher (6) has correlated saturated fatty acid critical temperatures with other physical properties, such as molar volume, normal boiling point and refractive indices.

Of the previous methods for estimating fatty acid densities, only Hammond and Lundberg's method (3) includes all commonly encountered fatty acids and incorporates the temperature dependency over a wide range. However, this method is not sufficiently accurate, giving errors of 2–5%.

Many correlations exist for the prediction of saturated liquid densities for a wide range of organic liquids, as reviewed by Spencer and Danner (7,8). One correlation that holds promise for application to the materials of interest here is the Rackett equation as modified by Spencer and Danner (7). The modified Rackett equation has been

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demonstrated to be effective with both hydrocarbons and nonhydrocarbons whose critical properties are known (7).

The original Rackett equation (9)

$$V_s = V_c Z_c^{(1-T_r)^{2/7}} \qquad [7]$$

and the modified Rackett equation

$$V_s = \left(\frac{RT_c}{P_c}\right) Z_{RA}^{[1+(1-T_r)^{2/7}]} \qquad [8]$$

are generalized equations that are used to estimate the molar volumes of saturated liquids, V_s. The critical constants, V_c, T_c, P_c, are the critical volume, temperature and pressure, respectively. T_r is the reduced temperature, R is the ideal gas constant and Z_{RA} is the Rackett parameter, a correlating parameter unique to each compound. The critical compressibility factor, Z_c, is always different from Z_{RA}.

The method of density estimation introduced here requires critical properties for the fatty acids. If the critical properties are not known, they must be estimated. The use of the modified Rackett equation also requires a single reference density for the fatty acid. The reference density can easily be located in most handbooks. The reference density must be for the liquid state. However, if a reference density is not available, a linear interpolation or extrapolation of fatty acid molar volumes at the same temperature and with the same degree of unsaturation can be used with some reduced accuracy.

Critical property estimation. The critical properties of fatty acids that thermally degrade must be estimated. There are several different ways in which this can be done. The group contribution method by Joback (cited in Ref. 10) is recommended for estimation of the critical pressure, P_c, where

$$P_c = (0.113 + 0.0032n_a - \sum \Delta_P)^{-2} \qquad [9]$$

Here, n_a is the number of atoms per molecule and Δ_P is the group contribution for critical pressure.

There are several different methods (Ambrose, Joback and Fedors, cited in Ref. 10) that can be used to estimate the critical temperature, T_c, as reviewed by Reid *et al.* (10). Fisher (11) has also derived several simple correlations based only upon the carbon chainlength, N_c, of which the most accurate is:

$$T_c = \frac{(N_c + 13.4)}{0.024984 + 0.00072433N_c} \qquad [10]$$

This equation agrees relatively well with published values but fails to take into account the effects of unsaturation. Therefore, for the sake of internal consistency, the chosen equation must compensate for the effect of the double bonds. The methods of Joback and Ambrose (cited in Ref. 10) both use group contribution and correction factors. Unfortunately, they require that the normal boiling point be known. Experimental T_b values have recently been determined by the Thermodynamic Research Center (TRC) at Texas A&M University (12) for the saturated fatty acids 1:0–20:0. However, there is no similar information available for the unsaturated fatty acids.

Joback (cited in Ref. 10) has an estimation for T_b that can be used to estimate T_c. However, this causes errors that average 7.49%, with some errors over 15%. These errors in T_b then lead to errors in T_c of at least 4%. The method of Ambrose (cited in Ref. 10) gives slightly better results, with average errors of 3%. The values of T_c obtained by these methods are shown in Table 1. The values are all calculated except for the values from Stephensen (13) and TRC (12), which are experimental values.

The Fedors group contribution method (see Ref. 10):

$$T_c = 535 \log \left(\sum \Delta_T\right) \qquad [11]$$

with critical-temperature group contribution, Δ_T, was used for critical temperature estimation in this work. This method was recommended by Reid *et al.* (10) when the normal boiling point is not available, which is quite often the case for the higher fatty acids and for the unsaturated fatty acids, as they generally decompose before they reach their normal boiling points. A comparison of T_c values, obtained by the Fedors method (cited in Ref. 10), gave average errors of less than 1% when compared to published literature values (12,13) of saturated fatty acids.

The Rackett parameter, Z_{RA}, is a unique correlating parameter for each compound. It can be estimated two ways. The first method solves the modified Rackett equation directly for Z_{RA} with a reference density, ϱ, at any given temperature:

$$Z_{RA} = \left[\frac{MW\, P_c}{\varrho\, R\, T_c}\right]^{[1+(1-T_r)^{2/7}]^{-1}} \qquad [12]$$

TABLE 1

Comparison of Estimated Critical Temperature Values

Fatty acid	T_c (°K)					
	Fedors	Ambrose	Joback	Eq. 10	Ref. 13	Ref. 12
1:0	551.15	579.05	574.78	560.13		
2:0	587.03	582.18	587.82	582.61	592.7	594.45
3:0	610.68	596.19	609.14	603.90	604.0	612.00
4:0	632.13	611.13	629.52	624.07	624.0	628.00
5:0	651.78	627.02	649.48	643.23	643.0	651.00
6:0	669.89	641.17	666.55	661.44	662.0	
7:0	686.69	652.37	679.75	678.77	679.0	
8:0	702.35	663.86	692.67	695.29	695.0	
9:0	717.03	674.49	704.29	711.04	711.0	
10:0	730.84	684.08	714.56	726.09	726.0	
11:0	743.87	693.96	725.00	740.48		
12:0	756.21	704.65	736.27	754.25		
13:0	767.92	714.39	746.59	767.44		
14:0	779.07	725.32	758.29	780.08		
15:0	789.72	734.85	768.73	792.21		
16:0	799.89	743.83	778.84	803.87		
16:1	800.34	853.50		803.87		
17:0	809.64	752.70	789.15	815.07		
18:0	819.00	762.86	801.17	825.84		
18:1	819.41	896.28		825.84		
18:2	819.82	902.40		825.84		
18:3	820.23	908.56		825.84		
19:0	827.99	771.10	811.58	836.21		
20:0	836.65	779.76	822.88	846.20		
20:1	837.03	939.50		846.20		
22:0	853.06	977.28		865.12		
22:1	853.41	983.07		865.12		
22:2	853.77	988.88		865.12		
24:0	868.38	1021.26		882.74		
24:1	868.71	1026.93		882.74		
26:0	882.76	1065.45		899.20		

Reference densities were taken from *Lange's Handbook of Chemistry* (14) and the *CRC Handbook of Chemistry and Physics* (15). The derived modified Rackett parameters for fatty acids are given in Table 2.

The second approach for developing the Rackett parameters uses a least-squares fitting of experimental values of density as a function of temperature. Unfortunately, limited data are available, so use of Equation 12 was necessary.

Fatty acid density estimation comparison. Of the previous fatty acid density estimation methods, only Equations 3 and 4 gave the versatility and temperature dependence needed for complete fatty acid density estimation. The modified Rackett equation (Equation 8), the Verschaffelt equation (Equation 5), and Equations 3 and 4 were compared to experimental data from the literature over a wide range of temperatures. Table 3 includes comparisons of unsaturated fatty acid densities (4,16), along with two unsaturated fatty acids (18:1 and 22:1) (17).

The modified Rackett equation predicts the liquid density of fatty acids over wide temperature ranges with an average error of 0.33%. It is more accurate than either Hammond and Lundberg's method or the Verschaffelt equation.

Vegetable oil density estimation. The most widely used density estimation method for vegetable oils was developed by Lund (18). The relation is:

$$sg\ (15/15°\mathrm{C}) = 0.8475 + 0.00030\ SV + 0.00014\ IV \qquad [13]$$

where sg is the specific gravity of the vegetable oil at 15°C compared to water at 15°C, SV is the saponification value and IV is the iodine value of the oil. Lund's equation is

TABLE 2

Rackett Parameters for Fatty Acids with Estimated T_c, P_c and Z_{RA} from a Reference Density

Fatty acid	T_c (°K)	P_c (bar)	Z_{RA}	Density reference
1:0	551.15	67.96	.2024	(14)
2:0	587.03	57.31	.2270	(14)
3:0	610.68	49.80	.2394	(14)
4:0	632.13	43.68	.2466	(14)
5:0	651.78	38.63	.2497	(14)
6:0	669.89	34.40	.2506	(14)
7:0	686.69	30.83	.2499	(14)
8:0	702.35	27.79	.2488	(14)
9:0	717.03	25.18	.2463	(14)
10:0	730.84	22.92	.2441	(14)
11:0	743.87	20.95	.2416	(14)
12:0	756.21	19.22	.2391	(15)
13:0	767.92	17.70	.2358	(est)
14:0	779.07	16.35	.2326	(14)
15:0	789.72	15.15	.2298	(14)
16:0	799.89	14.08	.2267	(14)
16:1	800.34	14.71	.2290	(est)
17:0	809.64	13.12	.2237	(15)
18:0	819.00	12.25	.2205	(14)
18:1	819.41	12.76	.2230	(14)
18:2	819.82	13.31	.2255	(14)
18:3	820.23	13.89	.2284	(14)
19:0	827.99	11.47	.2177	(est)
20:0	836.65	10.76	.2149	(14)
20:1	837.03	11.18	.2172	(est)
22:0	853.06	9.52	.2095	(14)
22:1	853.41	9.87	.2116	(14)
22:2	853.77	10.24	.2103	(est)
24:0	868.38	8.49	.2040	(est)
24:1	868.71	8.78	.2063	(est)
26:0	882.76	7.61	.1990	(est)

TABLE 3

Comparison of Experimental and Calculated Liquid Densities for Saturated Fatty Acids over Wide Temperature Ranges[a]

				Average percent absolute error when calculated by:		
Fatty acid	Temperature range (°C)	Density reference source	Number of points	Hammond and Lundberg[b]	Verschaffelt Equation[c]	Modified Rackett[d]
3:0	−36-119	(16)	18	2.75	0.36	0.39
4:0	0-260	(4)	14	5.26	0.33	0.76
4:0	0-92	(16)	16	0.90	1.11	0.12
5:0	−40-260	(4)	16	3.21	0.31	0.52
6:0	0-280	(4)	15	2.80	0.26	0.66
6:0	−20-101	(16)	12	0.37	0.57	0.37
8:0	20-300	(4)	15	1.60	0.23	0.36
8:0	16-124	(16)	13	0.34	0.71	0.28
10:0	40-300	(4)	14	1.46	0.17	0.50
10:0	32-140	(16)	9	0.88	1.37	0.07
12:0	60-300	(4)	13	2.12	0.29	0.34
12:0	42-136	(16)	7	1.99	0.36	0.08
14:0	60-300	(4)	13	2.84	0.16	0.35
14:0	57-149	(16)	11	1.90	0.54	0.26
16:0	80-300	(4)	12	3.78	0.09	0.37
16:0	64-149	(16)	11	2.36	0.64	0.59
17:0	91-147	(16)	6	3.20	—	0.26
18:0	80-300	(4)	12	4.54	0.11	0.32
18:0	70-145	(16)	9	2.99	0.37	0.10
18:1	24-110	(17)	8	—	—	0.10
22:1	38-110	(17)	7	—	—	0.08
			Average:	2.38	0.44	0.33

[a]Percent error = [(calc. − exp.)/exp.] * 100. [b]Reference 3, Equations 3 and 4. [c]Equation 5. [d]Equation 8.

accurate for a wide variety of oils. However, the saponification value test is becoming obsolete (19), and both tests are being replaced by the increasingly available and accurate techniques of gas and liquid chromatography for composition determination.

The SV and IV can also be calculated from the fatty acid composition of the oil. The calculations for the SV and IV are based on their definitions (19):

$$SV = \frac{56.104 \cdot 1000 \cdot 3}{MW_{oil}} \qquad [14]$$

$$IV = \frac{100 \cdot 126.9045 \cdot 2 \cdot 3 \cdot D}{MW_{oil}} \qquad [15]$$

where MW_{oil} is found with Equation 22 (presented later).

Paquot (20) notes that the change in density per °C is approximately −0.00068 for all vegetable oils. This provides a temperature dependence to use with Lund's equation (Equation 13) with SV and IV being determined by Equations 14 and 15.

Determination of liquid density for vegetable oils. Vegetable oil density is estimated by first computing the density of the liquid mixture of fatty acids and then including a correction factor to account for the triglyceride form. Density estimation for a liquid mixture as an extension of the modified Rackett equation was given by Spencer and Danner (21):

$$V_{s,\,mix} = R\left(\sum \frac{x_i\, T_{c_i}}{P_{c_i}}\right)\left(\sum x_i\, Z_{RA_i}\right)^{[1 + (1 - T_r)^{2/7}]} \qquad [16]$$

$$\varrho = \frac{\left(\sum x_i\, MW_i\right)}{V_{s,\,mix}} \qquad [17]$$

where x_i is the mole fraction of each component and $V_{s,\,mix}$ is the molar volume of the mixture.

Although a number of pseudocritical temperature rules exist for calculating the reduced temperature, a simple molar average of the critical temperatures (Kay Rule, Ref. 22) was used for the pseudocritical temperature, $T_{c,\,mix}$ in this work.

$$T_r = \frac{T}{T_{c,\,mix}} \qquad [18]$$

where

$$T_{c,\,mix} = \sum x_i\, T_{ci} \qquad [19]$$

A correction factor, F_c, is added to the calculated density to account for the triglyceride form of the fatty acids in the oils. The correction factor is independent of the temperature and depends solely on the oil type. The correction factor ranges from 0.02 to 0.04.

The correction factor was plotted *vs.* the molecular weight of the oil for a number of oils, including crambe, rapeseed, soybean, sunflower and corn oils. The correction factor equation for oils with molecular weight greater than 875 is:

$$F_c = 0.0236 + 0.000082\, |875 - MW_{oil}| \qquad [20]$$

For oils with molecular weight less than 875, the correction factor is:

$$F_c = 0.0236 + 0.000098\, |875 - MW_{oil}| \qquad [21]$$

where

$$MW_{oil} = 3\sum x_i\, MW_i + 38.0488 \qquad [22]$$

Thus, with Equation 17 and the correction factor from Equations 20 or 21, the density of a vegetable oil, ϱ_{oil}, is given by:

$$\varrho_{oil} = \frac{\left(\sum x_i\, MW_i\right)}{R\left(\sum \frac{x_i\, T_{c_i}}{P_{c_i}}\right)\left(\sum x_i\, Z_{RA_i}\right)^{[1 + (1 - T_r)^{2/7}]}} + F_c \qquad [23]$$

Vegetable oil density estimation comparison. The exact composition of a vegetable oil varies with climate, plant variety, geography, processing, *etc.* (19). A comparison of the two density estimation methods, the modified Rackett parameter and Lund's equation, is dependent upon the compositions. All densities and specific gravities were converted to apparent density, mass weighed in air per volume, for the comparison. Methods for the conversions were taken from References 14 and 15. Based upon the results shown in Table 4, the density estimations for vegetable oils from fatty acid compositions and the modified Rackett equation are slightly more accurate than the results from the Lund equation. Table 4 contains the vegetable oil density comparisons with densities and compositions from the same source (23–31), as well as the comparisons with the densities and compositions from different sources (32–42).

The modified Rackett equation presented here is the basis for a new method of estimating the density of vegetable oils from composition data. The resulting method can be easily incorporated into computer-aided process design programs that require the liquid density.

The modified Rackett equation is preferred over the use of other, simpler linear correlations such as those discussed by Costello and Bowden (4) and Fischer (6). Simple correlations, such as Equation 6, have several disadvantages. First, a new calculation sequence must be entered into the process simulator. Another disadvantage of relating densities to other physical properties is the large amount of experimental work that must be done. These additional data must then be placed into the component database library. The average process engineer has neither the time nor the programming expertise for an undertaking such as this. Thus, this approach does not lend itself to using commercial process simulators in the fatty acid and vegetable oil processing field.

However, the required critical properties and Rackett parameters are usually already available in the component database library or may be easily incorporated into the program data file by using values tabulated in this work without requiring that a new correlation be added to the thermodynamic calculation modules. The modified Rackett equation is also already present in the thermodynamic calculation module. Therefore, the modified Rackett equation can be used quickly and easily to calculate fatty acid and vegetable oil densities.

The modified Rackett equation gives accurate results for the densities of fatty acids and for vegetable oils, based on the fatty acid composition. Because the modified

TABLE 4

Comparison of Experimental and Calculated Vegetable Oil Liquid Densities

Type of oil	Density reference	Reference density temperature (°C)	Composition reference	Percent absolute error when calculated by: Lund[a]	Modified Rackett[b]
Crambe	(23)	25	(23)	0.43	0.04
Olive	(24)	25	(24)	0.03	0.14
Ouricurry palm kernel	(25)	25	(25)	0.04	0.04
Rapeseed	(26)	25	(26)	0.02	0.02
Safflower	(27)	25	(27)	0.06	0.08
Sesame	(28)	25	(28)	0.50	0.22
Soybean	(26)	25	(26)	0.03	0.01
Soybean	(29)	20	(29)	0.02	0.07
Soybean	(29)	30	(29)	0.03	0.13
Soybean	(29)	40	(29)	0.22	0.09
Span, rapeseed	(26)	25	(26)	0.03	0.00
Stillingia sylvatica	(30)	25	(30)	0.04	0.02
Sunflower	(31)	25	(31)	0.15	0.01
Tower, rapeseed	(26)	25	(26)	0.01	0.04
Babassu palm kernel	(1)	60	(32)	0.01	0.04
Coconut	(1)	25	(33)	0.04	0.19
Coconut	(1)	60	(33)	0.21	0.45
Corn	(1)	25	(34)	0.26	0.06
Cottonseed	(1)	15	(35)	0.06	0.19
Cottonseed	(1)	99	(35)	0.26	0.08
Cottonseed	(1)	200	(35)	0.64	0.68
Palm	(36)	25	(37)	0.08	0.17
Palm	(36)	75	(37)	0.14	0.26
Palm kernel	(1)	60	(38)	0.03	0.13
Peanut	(39)	25	(40)	0.04	0.10
Peanut	(39)	65	(40)	0.03	0.04
Peanut	(39)	76	(40)	0.17	0.13
Peanut	(39)	118	(40)	0.29	0.23
Peanut	(39)	156	(40)	0.51	0.52
Peanut	(39)	180	(40)	0.57	0.70
Soybean	(41)	25	(29)	0.12	0.03
Soybean	(41)	38	(29)	0.16	0.04
Soybean	(41)	50	(29)	0.16	0.08
Soybean	(42)	−10	(29)	0.09	0.15
Soybean	(42)	0	(29)	0.15	0.15
Soybean	(42)	10	(29)	0.12	0.07
Soybean	(42)	25	(29)	0.13	0.02
Soybean	(42)	40	(29)	0.15	0.01
			Overall average:	0.16	0.14

[a]Reference 18, Equation 13. [b]Reference 21.

Rackett equation uses critical properties that are either available (or predictable), it is ideally suited for use in computer-aided process design programs. The equation is computationally simple, requires readily available properties and does not need empirically-derived constants or equations.

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