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Refractive Index and Density of Acetone-Water Solutions

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Data for the density and refractive index of mixtures of acetone and water are presented. Deviations from ideality for such data are clearly shown if data are correlated with ideal volume per cent composition. If there is no deviation from ideality, density and refractive index may be exactly calculated from the ideal volume per cent composition. With deviation from ideality, a plot of refractive index vs. density in which the actual points are connected with the ideal points is very revealing. The slope of these tie lines may be related to the slopes corresponding to various specific refraction equations. In the case of the acetone-water blends, the simple Gladstone and Dale relation accurately represents the change of refractive index in relation to the change of density owing to the contraction on mixing. The possible effect of hydrogen bonding on the relation between refractive index and density for binary aqueous mixtures is pointed out. The presence of hydrogen bonding, or absence of it, may determine which specific refraction function best represents the data for a particular aqueous binary mixture.

THE study of data for binary mixtures of liquids, and of simple solutions of solids in liquids, is important for developing a better understanding of the liquid state (*5, 6, 20, 22, 23*). The properties of binary mixtures of liquids, particularly aqueous solutions, cannot be predicted with certainty from the properties of the pure materials but must be quantitatively evaluated from experimental data (*5, 6, 20, 22, 23*). This paper contributes data on binary mixtures of acetone and water and discusses briefly the application of refraction functions to the generalization of such data.

Data on binary mixtures of acetone and water were obtained some time ago for analytical use. These data have been checked recently in a second laboratory. These two sets of data are presented in Tables I and II. The composition data are recorded both as ideal volume % and weight %.

[1] Deceased.

Mole per cent composition is used in many discussions of binary mixtures (*19, 21, 22, 28*), but consideration from the volume % point of view still has some advantages. The space separating molecules is, of course, closely related to volume % composition.

Table III compares three sets of data at 50.26 ideal volume % water. The agreement among the three sets of data is satisfactory. Table IV compares the experimental density and refractive index data for the acetone and water used in this work with literature data. As the agreement is good, one may conclude that the materials used were of adequate purity.

Ideal volume % is the volume % calculated from the weight % assuming no volume change on mixing. By definition, density must plot linearly against ideal volume % if there is no volume change on mixing (*13, 19, 24, 25*), therefore the deviation from linearity when density is plotted against ideal volume % is a direct measure of deviation from

Table I. Sun Oil Experimental Data for Binary Mixtures of Acetone in Water [a](31)

Water % on Solution		Density, 20° C.	Spec. Grav., 60°/60° F.	Refractive Index		Refractivity Intercept $(n - d/2)$ 20° C.
Ideal vol. %	Wt. %			$n_D^{20° C.}$ ±0.00005	$n_D^{60° F.}$ ±0.00005	
0	0	0.7937	0.7995	1.35900	1.36163	0.9622
2	2.50	0.8011	0.8061	1.36024	1.36301	0.9597
5	6.21	0.8115	0.8179	1.36179	1.36430	0.9560
10	12.27	0.8287	0.8353	1.36389	1.36604	0.9496
15	18.17	0.8456	0.8513	1.36500	1.36718	0.9422
20	23.93	0.8604	0.8656	1.36568	1.36768	0.9355
25	27.55	0.8751	0.8796	1.36570	1.36752	0.9282
30	35.03	0.8894	0.8934	1.36557	1.36727	0.9209
40	45.62	0.9130	0.9166	1.36348	1.36519	0.9070
50	55.72	0.9335	0.9370	1.36085	1.36173	0.8941
60	65.37	0.9514	0.9543	1.35635	1.35752	0.8807
70	74.59	0.9654	0.9685	1.35107	1.35142	0.8684
80	83.42	0.9772	0.9799	1.34498	1.34557	0.8564
85	89.70	0.9828	0.9850	1.34202	1.34278	0.8506
90	91.89	0.9884	0.9899	1.33895	1.33948	0.8448
95	95.98	0.9937	0.9949	1.33591	1.33668	0.8391
98	98.42	0.9964	0.9975	1.33448	1.33476	0.8363
100	100.00	0.9986	1.0000	1.33322	1.33366	0.8339
(100%-0%)		+0.2049	+0.2005	-0.02578	-0.02797	-0.1292

[a] Volume % ignoring volume change on mixing—i.e., ideal volume % or Linear blend volume %.

Table II. University of Rhode Island Experimental Data for Binary Mixtures of Acetone in Water

Water		Density, G./Ml. 20° C.	Refractive Index, n_D^{20}	Refractivity Intercept, $n - d/2$, 20° C.	Density, G./Ml., 25° C.	Refractive Index, n_D^{25}	Refractivity Intercept, $n - d/2$, 25° C.
Ideal vol. %	Wt. %						
	100.00	0.99823	1.3330	0.8339	0.99707	1.3325	0.8340
87.80	90.08	0.9853	1.3402	0.8476	0.9837	1.3395	0.8477
75.72	70.74	0.9719	1.3472	0.8613	0.9695	1.3465	0.8618
64.24	69.39	0.9564	1.3540	0.8758	0.9535	1.3528	0.8761
53.99	59.68	0.9396	1.3585	0.8887	0.9360	1.3574	0.8894
44.11	49.89	0.9203	1.3624	0.9023	0.9167	1.3606	0.9023
35.90	41.41	0.9020	1.3642	0.9132	0.8974	1.3624	0.9137
24.57	29.13	0.8736	1.3654	0.9286	0.8690	1.3634	0.9288
16.06	19.45	0.8486	1.3647	0.9404	0.8474	1.3638	0.9401
7.86	9.72	0.8210	1.3624	0.9519	0.8167	1.3602	0.9519
Purified Acetone		0.7912	1.3586	0.9630	0.7853	1.3559	0.9633

Table III. Agreement Between Three Sets of Data for Acetone in Water

	Line	Water, Vol. %	Density	Refractive Index	Refractivity Intercept
SUN OIL DATA					
Wikingsson, 1938	A	50.0	0.9335	1.3590	0.8941
Wikingsson Interpolated	B	50.26	0.9340	1.3607	0.8937
Armes-Logan[a], 1963	C	50.26	0.9332	1.3604	0.8938
Difference	C-B		-0.0008	-0.0003	-0.0001
UNIVERSITY OF RHODE ISLAND DATA					
Bajwa-Lin, Table II, Interpolated	D	50.26	0.9323	1.3600	0.8938
Difference	C-D		+0.0009	+0.0004	0.0000

[a] Refractive indices were obtained with Bausch and Lomb Abbe refractometer. Densities were obtained with 10-ml. bicapillary pycnometers, ASTM D-941.

ideality. Where there is no volume change on mixing, refractive index is also additive on a volume % basis (2, 10, 12, 19, 24, 25). Therefore, if it is desired to emphasize the agreement with, or deviation from, ideality, it is desirable to plot density and refractive index against ideal volume % composition.

The data for Tables I and II at 20° C. have been plotted in this way in Figure 1. Note that refractive index goes through a maximum at about 25%, and that there is also substantial deviation from linearity for density, although no maximum appears.

Since density and refractive index are linearly additive on a volume % basis (8), it is also instructive to plot refractive index against density as has been done in Figure 2. In this graph, the solid curve represents the experimentally determined points. The dashed line connecting the points for acetone and water represents the densities and refractive indices which would have been observed if there had been no volume change on mixing. Dashed tie lines have been used to show the effect of volume change on mixing. Note that the tie lines are roughly parallel. The slope $\Delta n/\Delta d$ of these tie lines is 0.4 ± 0.1.

The equation for refractivity intercept (7, 9, 15, 16) is

$$r_i = n - 0.5d \qquad (1)$$

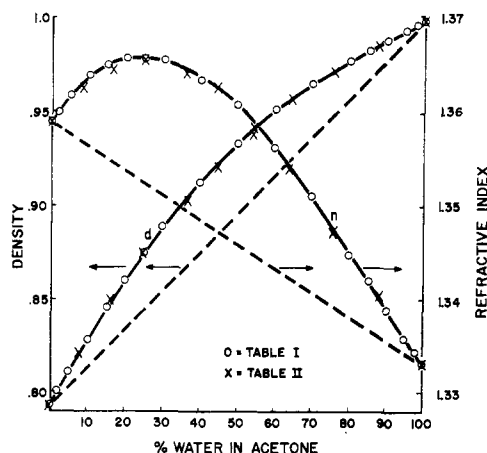where n = refractive index and d = density. Since the

Figure 1. Refractive index and density vs. ideal
volume % for acetone and water

Figure 2. Refractive index vs. density for
acetone and water

graphic slope (0.5) of this function is close to the observed slope of the tie lines in Figure 2, a plot of refractivity intercept *vs.* ideal volume % should be much closer to linear than the corresponding graphs for density or refractive index. Figure 3 shows such a plot for acetone and water which is, in fact, not far from linear. This was the basis for the recommendation that refractivity intercept be plotted against % composition (21).

The choice between ideal volume % and weight % for such plots depends upon the use for which the graphs are intended. In general, a high degree of linearity is achieved with the volume % plot (27). However, refractivity intercept is nearly linear with weight % for many binary mixtures (1, 21, 27). The deviation from linearity for various functions plotted against composition for a variety of binary aqueous blends will be discussed more fully in another communication (14).

In considering volume change and related effects, there is an appreciable amount of free volume or "expansion volume" in liquids. Traube (26) many years ago showed that there is a molecular increment associated with each dissolved unassociated molecule which he called "the molecular solution covolume." This molecular increment of volume has been verified by the study of the molecular volumes of unassociated molecules such as hydrocarbons and fluorinated hydrocarbons (9, 11, 12). At any one temperature or pressure, it is a constant independent of the molecular weight of the dissolved molecule. There is also free volume, or expansion volume, associated with the segments of large molecules. When volume is changed by temperature or pressure, or when there is volume change on mixing, one can think of it as a change in the free volume, since the bond lengths and bond distances in the molecules themselves do not change (7).

The slope $\Delta n/\Delta d$ of the tie lines (Figure 2) when density is changed is significant (15). Many specific refraction equations relating n to d have been published (3,4,15-18). The slope $\Delta n/\Delta d$ increases in order (15, p. 510-11) for the following specific refraction equations:

Newton (15, 16)

$$\frac{n^2 - 1}{d} = C_1 \tag{2}$$

Gladstone & Dale (4)

$$\frac{n - 1}{d} = C_2 \tag{3}$$

Eykman (3)

$$\left(\frac{n^2 - 1}{n + 0.4}\right)\frac{1}{d} = C_3 \tag{4}$$

Lorentz-Lorenz (17, 18)

$$\left(\frac{n^2 - 1}{n^2 + 2}\right)\frac{1}{d} = C_4 \tag{5}$$

The slope for such functions varies a little with the level of density and refractive index, but the order is always the same. Using the average value for the nine isomeric heptanes (15), namely, d = 0.6860 and n = 1.3880, we have the following values for $\Delta n/\Delta d$: Newton 0.485; Gladstone and Dale 0.57; Eykman 0.60; and Lorentz-Lorenz 0.64.

Table IV. Comparison of Observed and Literature Data
for Acetone and Water

	$d^{20 \cdot 4}$	n_D^{20}	$n - d/2$
ACETONE			
I.C.T. (30)	0.7915	1.3591	0.9634
1963 Univ. of Rhode Island	0.7912	1.3586	0.9633
1963 Sun Oil	0.7911	1.3587	0.9632
1938 Sun Oil	0.7937	1.3590	0.9622
WATER			
MCA (32)	0.99823[a]	1.33299[a]	0.8339
1963 Univ. of Rhode Island	0.99823[b]	1.3330[b]	0.8339
1963 Sun Oil	0.9980	1.3330[b]	0.8340
1938 Sun Oil	0.9986	1.33322	0.8339

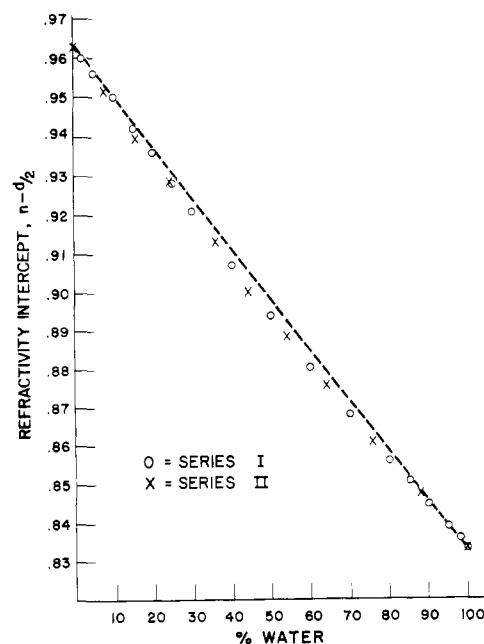[a] Rounded to 5 decimals. [b] Calibration value.

Figure 3. Refractivity intercept vs. ideal
volume % for acetone and water

The Eykman function accurately represents the effect of temperature and pressure on organic liquids (3, 7, 13, 15, 16, 29). The Lorentz-Lorenz function (15, 16) predicts too large a change of refractive index for a given change of density, and the Gladstone and Dale equation (4) predicts too little change of refraction. The plus deviation in refractive index of the Lorentz-Lorenz equation is almost exactly equal to the minus deviation of the Gladstone and Dale relation.

The Newton specific refraction has been shown to have a $\Delta n/\Delta d$ slope of about 15% less than the Gladstone and Dale equation. The Newton specific refraction has also been shown to represent quite well the relation between n and d for groups of hydrocarbon isomers at constant temperature and pressure (7, 13, 15, 16, 29). Since the Newton specific refraction and the Sellmeier-Drude dispersion equation give the same relation between refractive index and density, one can say that if the number and frequency of the dispersion electrons are not changed when the density is changed that the Newton equation will apply. It is therefore of interest to see how nearly the slope $\Delta n/\Delta d$ associated with the volume change on mixing for acetone and water agrees with those specific refraction equations.

A preliminary examination indicated that the Gladstone and Dale function would come close to agreeing with the data for the volume change on mixing effect with water and acetone. If the Gladstone and Dale relation applies, several simple relations are true, namely:

$$\Delta n = (n_L - 1)(d_r - 1) \quad (6)$$

$$\Delta r_i = (r_{i_L} - 1)(d_r - 1) \quad (7)$$

$$d_r = d/d_L \quad (8)$$

In these equations Δn and Δr_i are the increment of refractive index and refractivity intercept observed by subtracting from the observed values the values calculated for the ideal volume % blend; d_r is the experimental density (d) divided by the density (d_L) calculated for the ideal volume % blend—i.e., by linear interpolation on an ideal volume % basis; n_L = the refractive index calculated for the ideal volume % blend by linear interpolation on a volume % basis.

It can easily be shown that if the above equations apply

$$\Delta n/\Delta d = \frac{n_L - 1}{d_L} \quad (9)$$

In Table V, values are given for d_L, d_r, n_L, $(n_L - 1)/d_L$ (which is the calculated slope), $\Delta n/\Delta d$ the observed slope, and the ratio of the calculated and observed slope. These data show that for the specific case of acetone water mixtures, the Gladstone and Dale relation can be used to calculate either the slope of the tie lines in Figure 2 or the absolute magnitude of Δn. Table VI shows the agreement in terms of the calculated Δn and the difference between this and the observed Δn.

The Gladstone and Dale function and the related equations apply quite well for the refractive index density relations associated with volume change on mixing of acetone and water. Kurtz, Thompson, and Camin have shown (14) for many other binary aqueous solutions of organic liquids that Newton specific refraction, which has a slope about 15% less than the Gladstone and Dale equation, represents the relation between refractive index and density better than that equation. There is some indication

Table V. Comparison of Calculated and Observed $\Delta n/\Delta d$ for Blends of Water and Acetone Using Gladstone and Dale Specific Refraction

Vol. % Water in Acetone[a]		d_L[b]	Density Ratio, d_r (d/d_L)	Volume Decrease, %	n_L[c]	($n_L - 1$[c]/d_L)	($\Delta n/\Delta d$), Observed	Ratio, F/G
Series I	Series II							
A		B	C	D	E	F	G	H
2		0.7978	1.0041	0.4	1.3585	0.4494	0.533	0.843
5		0.8040	1.0094	0.9	1.3577	0.4449	0.540	0.824
	7.86	0.8095	1.0167	1.6	1.3566	0.4405	0.429	1.027
10		0.8142	1.0178	1.8	1.3564	0.4377	0.514	0.852
15		0.8244	1.0257	2.5	1.3551	0.4307	0.466	0.924
	16.06	0.8244	1.0294	2.9	1.3549	0.4304	0.421	1.022
20		0.8346	1.0308	3.0	1.3538	0.4239	0.467	0.908
	24.57	0.8422	1.0373	3.6	1.3523	0.4183	0.417	1.003
25		0.8459	1.0345	3.3	1.3526	0.4168	0.450	0.926
30		0.8552	1.0400	3.8	1.3513	0.4108	0.417	0.985
	35.90	0.8655	1.0422	4.0	1.3494	0.4037	0.405	0.997
40		0.8757	1.0426	4.1	1.3487	0.3982	0.406	0.981
	44.11	0.8825	1.0428	4.1	1.3473	0.3935	0.399	0.986
50		0.8962	1.0416	4.0	1.3461	0.3862	0.394	0.980
	53.99	0.9030	1.0405	3.9	1.3448	0.3818	0.402	0.950
60		0.9166	1.0379	3.7	1.3435	0.3748	0.368	1.018
	64.24	0.9243	1.0347	3.4	1.3422	0.3702	0.367	1.009
70		0.9371	1.0302	2.9	1.3410	0.3639	0.358	1.016
	75.72	0.9480	1.0252	2.5	1.3392	0.3578	0.335	1.068
80		0.9576	1.0204	2.0	1.3384	0.3534	0.337	1.049
85		0.9678	1.0155	1.5	1.3371	0.3483	0.327	1.085
	87.80	0.9730	1.0127	1.3	1.3361	0.3454	0.333	1.037
90		0.9781	1.0105	1.0	1.3358	0.3433	0.306	1.122
95		0.9884	1.0054	0.5	1.3345	0.3384	0.262	1.292
98		0.9945	1.0019	0.2	1.3337	0.3355	0.389	0.863

Average of 16, series I	0.978
Average of 9 , series II	1.014
Average of 25, both series	0.991
Standard deviation series I	0.119
Standard deviation series II	0.036
Standard deviation both series	0.099
Standard deviation both series omitting one point	0.072

[a] Ideal volume %, that is on no-volume change basis. [b] The symbol d_L is used for the density calculated by linear interpolation that is on the basis of no volume change on mixing. The ratio d/d_L shows the actual density divided by the ideal density. [c] The symbol n_L is used for the refractive index calculated by linear interpolation on the ideal volume % basis.

Table VI. Comparison of Calculated and Observed Refractivity Indices of Mixtures[a]

Ideal Vol. % Water	$10^4 \times$ Refractive Index Increment		$10^4 \times \Delta$ Refractive Index	
	Calculated from volume change[2]	Observed	Table I data	Table II data
2	0.0015	0.0017	+2	
5	0.0034	0.0041	+7	
7.86	0.0060	0.0058		−2
10	0.0063	0.0075	+12	
15	0.0091	0.0099	+8	
16.06	0.0104	0.0102		−2
20	0.0109	0.0110	+10	
24.57	0.0131	0.0131		0
25	0.0122	0.0131	+9	
30	0.0141	0.0143	+2	
35.90	0.0147	0.0148		+1
40	0.0147	0.0148	+1	
44.11	0.0149	0.0151		+2
50	0.0144	0.0148	+4	
53.99	0.0140	0.0137		−3
60	0.0130	0.0129	−1	
64.24	0.0119	0.0118		−1
70.0	0.0103	0.0101	−2	
75.72	0.0086	0.0080		−6
80.0	0.0069	0.0066	−2	
85	0.0052	0.0049	−3	
87.80	0.0043	0.0041		−2
90	0.0035	0.0032	−3	
95	0.0018	0.0014	−4	
98	0.0006	0.0008	+2	
	Average deviation:		4.5	2.1
	Deviation of average:		+2.6	−1.4

[a] Refractive index increment $\Delta n = (n_L - 1)(d_r - 1)$, see Table V.

that the Gladstone and Dale function will represent data well when there is mild hydrogen bonding, as with acetone and water, and that the Newton specific refraction will apply when there is no hydrogen bonding (14). The study of more aqueous binary solutions should clarify this point.

ACKNOWLEDGMENT

The authors acknowledge the assistance of C. T. Armes and S. A. Logan of Sun Oil Co. for confirmatory physical property data shown in Table III and to M. A. Bajwa and K. K. Lin of the University of Rhode Island for the data presented in Table II.

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