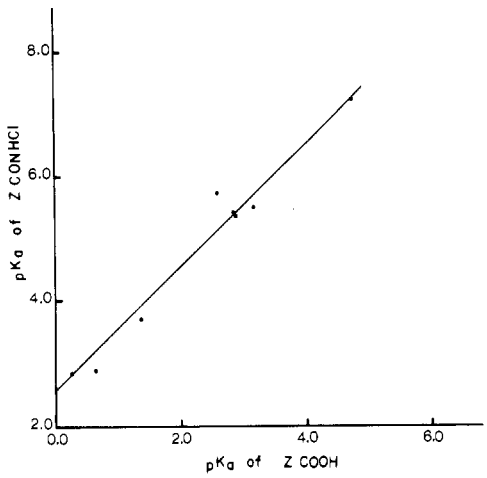


**Figure 1.** p$K_a$ values of ZCONHCl vs. p$K_a$ values of ZCOOH in water at 25 °C.

### Experimental Section

The *N*-haloamides were prepared by the sodium hypohalite method (*1*) except for *N*-fluorourethane (*3*).

The potentiometric titrations were carried out on a 0.01 M solution at 25 °C using an Orion-type pH meter Model 801, a Sargent-Welch glass electrode type S-30050-15, and a saturated calomel electrode type S-30080-15. The buret, containing an NaOH 0.01 M solution, had a digital syringe, Dosimat type, Metrohm Herisan Model E 412. Before and after each titration, the electrodes were calibrated using two buffers—lithium oxalate–oxalic acid and lithium succinate–succinic acid (*5*)—prepared in the same solvent as the titrated compound. The reproducibility was of 0.02 pH unit. The p$K_a$s of benzoic acid and *p*-nitrophenol determined by this method were found to be 5.68 and 7.86, respectively, in agreement with the literature values (*5, 7*).

### Acknowledgment

Our thanks are due to F. Comtois and M. Paquette for technical assistance.

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Received for review June 13, 1977. Accepted September 28, 1977. Financial assistance from the "Ministère de l'Education du Québec" and the National Research Council of Canada is gratefully acknowledged.

# Densities of Dilute Aqueous Solutions of Selected Ethers†

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**The densities of dilute aqueous solutions of 11 ethers [oxetane, oxepane, 1,3-dioxane, 1,3-dioxepane, 1,3,5-trioxane, 2,4,6-trimethyl-1,3,5-trioxane (paraldehyde), 2,5-dimethoxytetrahydrofuran, dimethoxymethane, diethoxymethane, 1,2-dimethoxyethane, 1,2-diethoxyethane] have been determined at 25 °C by means of a differential buoyancy technique. The solute apparent molar volumes and excess molar volumes are presented. An empirical equation is proposed to evaluate the limiting partial molar volumes of cyclic ethers with the formula $(CH_2)_nO_m$.**

The volumetric properties of nonelectrolyte aqueous solutions, in conjunction with other thermodynamic properties, provide useful information about solute–water interactions (*3*). In continuation of previous studies on the volumetric behavior of ethers in water (*1*), we have determined the densities at 25 °C of dilute aqueous solutions of some cyclic monoethers (oxetane, oxepane), cyclic di- and triethers (1,3-dioxane, 1,3-dioxepane, 1,3,5-trioxane, 2,4,6-trimethyl-1,3,5-trioxane or paraldehyde, 2-5-dimethoxytetrahydrofuran), and open-chain diethers (dimethoxymethane, diethoxymethane, 1,2-dimethoxyethane, 1,2-diethoxyethane). From density data, the solute apparent molar volumes have been calculated at each concentration, and the results have been extrapolated to give the partial molar volumes at infinite dilution. These limiting quantities, in conjunction with the known molar volumes of pure liquid compounds, enabled the calculation of excess molar volumes.

† Partly communicated at the meeting of the Società Chimica Italiana, Pisa, Dec 1975. *Chim. Ind. (Milan)*, **58**, 221 (1976).

### Experimental Section

All chemicals were commercial products of the highest available purity. Solid 1,3,5-trioxane was sublimed under vacuum on a cold finger in a closed glass apparatus. The liquid compounds were fractionally distilled, at atmospheric pressure, after prolonged reflux over metallic sodium, using a Todd column with 30 theoretical plates. Oxepane was further rectified by a Perkin-Elmer 251 spinning band column. All the samples used in the experiments showed a GLC purity ≥99.8% and a water content less than 0.1%, as measured by a Karl Fischer reagent. In the case of 2,5-dimethoxytetrahydrofuran, the cis–trans

0021-9568/78/1723-0065$01.00/0 

**Table I. Densities and Apparent Molar Volumes of Ethers in Aqueous Solutions at 25 °C**

| Concen, mol $L^{-1}$ | Density, g $cm^{-3}$ | $\phi_v$, $cm^3$ $mol^{-1}$ |
|---|---|---|
| | Oxetane | |
| 0.0213 | 0.996 982 | 61.20 |
| 0.0285 | 0.996 956 | 61.37 |
| 0.0367 | 0.996 929 | 61.41 |
| 0.0493 | 0.996 896 | 61.27 |
| 0.0604 | 0.996 861 | 61.32 |
| 0.1087 | 0.996 718 | 61.27 |
| 0.1108 | 0.996 708 | 61.30 |
| 0.1116 | 0.996 699 | 61.37 |
| 0.2133 | 0.996 407 | 61.25 |
| 0.2146 | 0.996 395 | 61.29 |
| 0.2397 | 0.996 330 | 61.24 |
| 0.3816 | 0.995 927 | 61.19 |
| 0.4024 | 0.995 868 | 61.19 |
| 0.5248 | 0.995 549 | 61.11 |
| 0.6138 | 0.995 309 | 61.09 |
| 0.6766 | 0.995 146 | 61.07 |
| 0.7758 | 0.994 911 | 61.01 |
| 0.8756 | 0.994 670 | 60.97 |
| 0.9521 | 0.994 493 | 60.94 |
| | Oxepane | |
| 0.0156 | 0.996 968 | 105.40 |
| 0.0197 | 0.996 949 | 105.36 |
| 0.0244 | 0.996 927 | 105.30 |
| 0.0415 | 0.996 840 | 105.40 |
| 0.0560 | 0.996 774 | 105.31 |
| 0.0585 | 0.996 763 | 105.29 |
| 0.0868 | 0.996 630 | 105.25 |
| 0.1171 | 0.996 496 | 105.16 |
| 0.1185 | 0.996 488 | 105.17 |
| 0.1475 | 0.996 363 | 105.09 |
| 0.1840 | 0.996 220 | 104.96 |
| 0.2145 | 0.996 089 | 104.92 |
| | 1,3-Dioxane | |
| 0.0179 | 0.997 180 | 80.82 |
| 0.0373 | 0.997 324 | 80.86 |
| 0.0387 | 0.997 336 | 80.81 |
| 0.0782 | 0.997 638 | 80.77 |
| 0.1123 | 0.997 893 | 80.80 |
| 0.1586 | 0.998 247 | 80.77 |
| 0.2547 | 0.998 980 | 80.75 |
| 0.2587 | 0.999 013 | 80.74 |
| 0.4379 | 1.000 393 | 80.70 |
| 0.4540 | 1.000 526 | 80.68 |
| 0.5649 | 1.001 387 | 80.66 |
| | 1,3-Dioxepane | |
| 0.0235 | 0.997 197 | 95.93 |
| 0.0258 | 0.997 212 | 95.95 |
| 0.0265 | 0.997 219 | 95.85 |
| 0.0506 | 0.997 375 | 95.90 |
| 0.0622 | 0.997 444 | 95.99 |
| 0.0817 | 0.997 574 | 95.94 |
| 0.1116 | 0.997 769 | 95.93 |
| 0.1325 | 0.997 911 | 95.87 |
| 0.1422 | 0.997 967 | 95.93 |
| 0.1995 | 0.998 357 | 95.84 |
| 0.2400 | 0.998 628 | 95.82 |
| 0.3055 | 0.999 076 | 95.76 |
| 0.4118 | 0.999 805 | 95.71 |
| 0.4772 | 1.000 267 | 95.66 |
| 0.5839 | 1.001 018 | 95.61 |
| 0.7495 | 1.002 197 | 95.54 |
| 0.8673 | 1.003 040 | 95.50 |
| | 1,3,5-Trioxane | |
| 0.0088 | 0.997 229 | 69.50 |
| 0.0180 | 0.997 421 | 69.45 |
| 0.0330 | 0.997 729 | 69.54 |
| 0.0422 | 0.997 921 | 69.55 |
| 0.0647 | 0.998 386 | 69.55 |
| 0.0893 | 0.998 897 | 69.54 |
| 0.1033 | 0.999 189 | 69.53 |
| 0.1324 | 0.999 789 | 69.56 |
| 0.1735 | 1.000 638 | 69.57 |
| 0.2084 | 1.001 360 | 69.58 |
| 0.2144 | 1.001 484 | 69.58 |
| 0.2489 | 1.002 195 | 69.60 |
| 0.2500 | 1.002 227 | 69.56 |
| 0.4850 | 1.007 068 | 69.62 |
| 0.5032 | 1.007 457 | 69.59 |
| 0.6994 | 1.011 436 | 69.71 |
| 0.7133 | 1.011 771 | 69.64 |
| | Paraldehyde | |
| 0.0213 | 0.997 235 | 123.59 |
| 0.0243 | 0.997 259 | 123.69 |
| 0.0467 | 0.997 461 | 123.62 |
| 0.0488 | 0.997 479 | 123.62 |
| 0.0492 | 0.997 481 | 123.65 |
| 0.0509 | 0.997 497 | 123.64 |
| 0.0867 | 0.997 816 | 123.63 |
| 0.0941 | 0.997 885 | 123.59 |
| 0.1003 | 0.997 945 | 123.55 |
| 0.1353 | 0.998 262 | 123.53 |
| 0.1637 | 0.998 529 | 123.46 |
| 0.2103 | 0.998 966 | 123.39 |
| 0.2180 | 0.999 036 | 123.30 |
| 0.2711 | 0.999 545 | 123.30 |
| 0.3092 | 0.999 904 | 123.27 |
| 0.3685 | 1.000 492 | 123.17 |
| 0.4235 | 1.001 030 | 123.11 |
| 0.4419 | 1.001 217 | 123.08 |
| 0.5100 | 1.001 894 | 123.01 |
| 0.5636 | 1.002 431 | 122.97 |
| 0.6281 | 1.003 094 | 122.89 |
| 0.6544 | 1.003 372 | 122.85 |
| 0.7004 | 1.003 833 | 122.83 |
| 0.7211 | 1.004 031 | 122.83 |
| | 2,5-Dimethoxytetrahydrofuran | |
| 0.0130 | 0.997 177 | 122.32 |
| 0.0234 | 0.997 284 | 122.33 |
| 0.0276 | 0.997 326 | 122.33 |
| 0.0524 | 0.997 583 | 122.27 |
| 0.0527 | 0.997 588 | 122.22 |
| 0.0903 | 0.997 976 | 122.22 |
| 0.0983 | 0.998 064 | 122.15 |
| 0.1433 | 0.998 538 | 122.10 |
| 0.1594 | 0.998 704 | 122.11 |
| 0.2122 | 0.999 273 | 122.03 |
| 0.2838 | 1.000 041 | 121.96 |
| 0.4259 | 1.001 614 | 121.79 |
| 0.5539 | 1.003 058 | 121.67 |
| 0.6937 | 1.004 666 | 121.53 |
| 0.8193 | 1.006 122 | 121.44 |
| | Dimethoxymethane | |
| 0.0314 | 0.996 917 | 80.42 |
| 0.0385 | 0.996 888 | 80.41 |
| 0.0838 | 0.996 706 | 80.38 |
| 0.0850 | 0.996 699 | 80.41 |
| 0.1784 | 0.996 328 | 80.35 |
| 0.2130 | 0.996 191 | 80.35 |
| 0.3510 | 0.995 659 | 80.28 |
| 0.4069 | 0.995 437 | 80.29 |
| 0.6219 | 0.994 643 | 80.20 |
| 0.7349 | 0.994 236 | 80.16 |
| 0.9126 | 0.993 604 | 80.10 |
| 1.0529 | 0.993 104 | 80.08 |
| 1.2317 | 0.992 478 | 80.04 |

**Table I** *(continued)*

| Concen, mol $L^{-1}$ | Density, g $cm^{-3}$ | $\phi_v$, $cm^3$ $mol^{-1}$ | Concen, mol $L^{-1}$ | Density, g $cm^{-3}$ | $\phi_v$, $cm^3$ $mol^{-1}$ |
|---|---|---|---|---|---|
| | Diethoxymethane | | 0.2723 | 0.995 665 | 95.47 |
| 0.0099 | 0.996 955 | 113.60 | 0.2728 | 0.995 653 | 95.51 |
| 0.0127 | 0.996 923 | 114.05 | 0.3279 | 0.995 401 | 95.42 |
| 0.0222 | 0.996 840 | 113.69 | 0.3754 | 0.995 158 | 95.43 |
| 0.0288 | 0.996 772 | 113.97 | 0.4263 | 0.994 950 | 95.32 |
| 0.0292 | 0.996 773 | 113.79 | 0.5309 | 0.994 454 | 95.28 |
| 0.0554 | 0.996 526 | 113.85 | 0.5937 | 0.994 235 | 95.13 |
| 0.0604 | 0.996 487 | 113.72 | 0.7191 | 0.993 708 | 95.04 |
| 0.1013 | 0.996 104 | 113.78 | 0.8306 | 0.993 312 | 94.90 |
| 0.1126 | 0.996 000 | 113.77 | 0.8481 | 0.993 235 | 94.89 |
| 0.1354 | 0.995 800 | 113.67 | 0.8579 | 0.993 213 | 94.87 |
| 0.2093 | 0.995 125 | 113.66 | 1.0868 | 0.992 388 | 94.68 |
| 0.2377 | 0.994 869 | 113.64 | | | |
| 0.2668 | 0.994 633 | 113.52 | | 1,2-Diethoxyethane | |
| 0.3514 | 0.993 893 | 113.45 | 0.0202 | 0.996 871 | 127.17 |
| 0.3841 | 0.993 625 | 113.39 | 0.0315 | 0.996 771 | 127.25 |
| 0.4048 | 0.993 466 | 113.33 | 0.0353 | 0.996 738 | 127.24 |
| | | | 0.0598 | 0.996 532 | 127.12 |
| | 1,2-Dimethoxyethane | | 0.0862 | 0.996 309 | 127.09 |
| 0.0343 | 0.996 862 | 95.73 | 0.1349 | 0.995 916 | 126.93 |
| 0.0434 | 0.996 806 | 95.91 | 0.1529 | 0.995 769 | 126.90 |
| 0.0752 | 0.996 640 | 95.78 | 0.2107 | 0.995 316 | 126.76 |
| 0.1072 | 0.996 478 | 95.70 | 0.2634 | 0.994 925 | 126.60 |
| 0.1148 | 0.996 439 | 95.68 | 0.3423 | 0.994 349 | 126.42 |
| 0.1230 | 0.996 390 | 95.73 | 0.3888 | 0.994 029 | 126.31 |
| 0.1711 | 0.996 143 | 95.67 | 0.4625 | 0.993 527 | 126.15 |
| 0.1791 | 0.996 095 | 95.71 | 0.6615 | 0.992 252 | 125.79 |

**Table II. Volumetric Properties of Selected Ethers in Water at 25 °C**[a]

| Substance | $\bar{V}_2^\circ$, $cm^3$ $mol^{-2}$ | $h$,[b] $cm^3$ L $mol^{-2}$ | $V_2$, $cm^3$ $mol^{-1}$ | $\bar{V}^{\circ E}$, $cm^3$ $mol^{-1}$ |
|---|---|---|---|---|
| Oxetane | 61.35 ± 0.01 | −0.43 ± 0.02 | 65.0 | −3.7 |
| Oxepane | 105.46 ± 0.02 | −2.55 ± 0.17 | 113 | −7.6 |
| 1,3-Dioxane | 80.82 ± 0.02 | −0.30 ± 0.06 | 85.9 | −5.1 |
| 1,3-Dioxepane | 95.98 ± 0.02 | −0.66 ± 0.10 | 102.1 | −6.1 |
| 1,3,5-Trioxane | 69.51 ± 0.04 | 0.22 ± 0.10 | 74 | −4.5 |
| Paraldehyde | 123.71 ± 0.02 | −1.50 ± 0.13 | 134.0 | −10.3 |
| 2,5-Dimethoxytetrahydrofuran | 122.34 ± 0.04 | −1.50 ± 0.32 | 130 | −7.7 |
| Dimethoxymethane | 80.42 ± 0.02 | −0.36 ± 0.03 | 89.2 | −8.8 |
| Diethoxymethane | 113.88 ± 0.03 | −1.27 ± 0.10 | 126.3 | −12.4 |
| 1,2-Dimethoxyethane | 95.88 ± 0.03 | −1.37 ± 0.10 | 104.7 | −8.8 |
| 1,2-Diethoxyethane | 127.29 ± 0.03 | −2.55 ± 0.12 | 139.9 | −12.6 |
| Tetrahydrofuran[c] | 76.85 | −1.08 | 81.7 | −4.9 |
| Tetrahydropyran[c] | 91.73 | −1.70 | 98.2 | −6.5 |
| 1,3-Dioxolane[c] | 65.37 | 0.17 | 70.3 | −5.0 |
| 1,4-Dioxane[c] | 80.96 | −0.17 | 85.8 | −4.8 |

[a] The $\phi_v$ data were fitted to eq 3 by means of a weighted least-squares adjustment. The reported uncertainties are standard deviations. [b] In the case of the following ethers: 1,3-dioxepane (0.4), paraldehyde (0.3), 2,5-dimethoxytetrahydrofuran (0.2), dimethoxymetane (0.2), 1,2-dimethoxyethane (0.4), 1,2-diethoxyethane (0.4); the plot of $\phi_v$ vs. $C$ was found to be linear up to the concentration value indicated in parentheses. [c] Reference 1.

equimolar mixture was employed. Water used as solvent was deionized and then distilled from alkaline $KMnO_4$ solution.

The hydrostatic differential balance used for density measurements (with a precision of 1 ppm) and details of the apparatus and its method of operation have been described elsewhere (*1*). The solutions to be studied were prepared directly in the measuring vessel by successive additions of a weighed quantity of ether to a known quantity of water. Temperature oscillations inside the measuring vessel were within ±0.001 °C.

The density values, $d$, were calculated from the buoyancy value, $W$, expressed as a mass difference, by means of the following equation

$$d = d_0 + W/V \qquad (1)$$

where $V$ is the exactly known volume of the float and $d_0$ the density of pure water at 25 °C ($d_0$ = 0.997 044 9 g $cm^{-3}$) (*4*).

## Results

Densities and apparent molar volumes, $\phi_v$, of the ethers are presented in Table I. The $\phi_v$ values were calculated from the experimental densities by use of the relation

$$\phi_v = \frac{M}{d_0} - \left(\frac{d}{d_0} - 1\right)\frac{1000}{C} \qquad (2)$$

where $M$ is the solute molecular weight and $C$ is its molar concentration. At least two series of determinations were carried out for each compound.

In Table II are collected data on the limiting partial molar volumes, $\bar{V}_2^\circ$, and the slope, $h$, of the straight line

$$\phi_v = \bar{V}_2^\circ + hC \qquad (3)$$

that represents the concentration dependence of $\phi_v$ in dilute solution. To facilitate comparison, data of related compounds

have been included in the table. Also reported are values of the excess partial molar volume at infinite dilution, $\bar{V}^{oE}$, calculated by

$$\bar{V}^{oE} = \bar{V}_2^{o} - V_2 \quad (4)$$

The molar volumes, $V_2$, of the pure liquids were evaluated from the average of density values available in the literature.

As far as we know, with the exception of $\bar{V}_2^{o}$ (25 °C) = 95.6 mL mol$^{-1}$ (*5*) for 1,2-dimethoxyethane, no density data are available with which the results presented here can be directly compared. The agreement between the $\bar{V}_2^{o}$ value of Terasawa et al. (*5*) determined dilatometrically and that obtained in this study by density measurements is reasonable.

## Discussion

***Partial Molar Volumes.*** From Table II, the effect on $\bar{V}_2^{o}$ of the chain lengthening appears evident. The $\bar{V}_2^{o}$ increment per methylene unit in homologous series, although dependent to a certain extent on molecular size, has the following average values: $\bar{V}_2^{o}(CH_2)$ = 14.7 cm$^3$ mol$^{-1}$ for cyclic monoethers, $\bar{V}_2^{o}(CH_2)$ = 15.3 cm$^3$ mol$^{-1}$ for cyclic diethers, $\bar{V}_2^{o}(CH_2)$ = 16.2 cm$^3$ mol$^{-1}$ for open-chain diethers.

As far as the effect of the number of oxygen atoms on $\bar{V}_2^{o}$ is concerned, the progressive introduction of oxygen atoms in cyclic ethers with a fixed number of carbon atoms (e.g., the changes oxetane → 1,3-dioxolane → 1,3,5-trioxane) is accompanied by a nearly constant increment in $\bar{V}_2^{o}$ (4.1 cm$^3$ mol$^{-1}$). This quantity, which appears surprisingly independent of both molecular size and relative positions of oxygen atoms in the molecule, can be identified with $\bar{V}_2^{o}(O)$—the ethereal oxygen contribution to partial molar volume.

Hence, there is some evidence that for cyclic ethers of the type $(CH_2)_nO_m$ there is a certain volume associated with each group in the molecule, so that $\bar{V}_2^{o}$ can be calculated by summing these volumes in a manner similar to that of Traube (*6*). A least-squares method led to the following empirical equation

$$\bar{V}_2^{*} = A + n\bar{V}_2^{o}(CH_2) + m\bar{V}_2^{o}(O) \quad (5)$$

where $\bar{V}_2^{*}$ is the calculated value of the limiting partial molar volume, $A$ = 12.7 cm$^3$ mol$^{-1}$ is a constant term, $\bar{V}_2^{o}(CH_2)$ = 14.9 cm$^3$ mol$^{-1}$, and $\bar{V}_2^{o}(O)$ = 4.1 cm$^3$ mol$^{-1}$. The $\bar{V}_2^{*}$ values calculated from eq 5 agree with the experimental $\bar{V}_2^{o}$ values of the nine $(CH_2)_nO_m$ ethers in Table II, with a standard deviation of ±0.4 cm$^3$ mol$^{-1}$.

Finally, one can observe that the open-chain diethers have always a greater $\bar{V}_2^{o}$ with respect to the corresponding cyclic diethers. The $\bar{V}_2^{o}$ increment in going from cyclic to open-chain compounds increases with increasing molecular dimensions.

***Concentration Dependence of the $\phi_v$.*** For all substances examined, the $\phi_v$ values show, in sufficiently dilute solution ($C$ < 0.2 mol L$^{-1}$), a linear dependence on the concentration. The value of the slope, $h$, is always negative, except for the 1,3,5-trioxane, and increases numerically within an homologous series, when the number of carbon atoms is increased.

The $h$ slopes and the $\bar{V}_2^{o}$ values are correlated with each other, and the correlation curve of monoethers is different from that of diethers and triethers. This different behavior between mono- and polyethers is analogous to that already observed for other classes of mono- and polyfunctional solutes (*2*).

***Excess Partial Molar Volumes.*** The transfer from the liquid state to the aqueous dilute solution produces always a volume contraction ($\bar{V}^{oE}$ < 0), which, in an homologous series, increases with chain lengthening. Moreover, for compounds with the same molecular size, the volume contraction diminishes as the oxygen number in the molecule increases (e.g., the $\bar{V}^{oE}$ trends in the series tetrahydropyran → 1,3-dioxane → 1,3,5-trioxane) and in going from open-chain to cyclic compounds.

***Note Added during the Revision.*** Shortly after this manuscript had been submitted, Edward et al. (Edward, J. T., Farrell, P. G., Shahidi, F., *J. Chem. Soc., Faraday Trans. 1*, **73**, 705 (1977)), published $\bar{V}_2^{o}$ values for seven of the ethers here considered. In general, their data agree within 0.2 cm$^3$ mol$^{-1}$ with ours.

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Received for review April 25, 1977. Accepted September 19, 1977. We are indebted to the Consiglio Nazionale delle Ricerche (Rome) for support of this work.