# Solubility of Krypton in Water and Seawater

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**The solubility of krypton in distilled water and seawater has been measured microgasometrically over the range 0–40 °C. The data have been corrected for nonideality and are fitted to equations in temperature and salinity of the form used previously to fit the solubilities of other gases. The fitted values have a precision of 0.2% and an estimated accuracy of 0.4% and are given in units of the Bunsen solubility coefficient as well as the solubility in milliliters/liter and milliliters/kilogram from moist air at 1 atm total pressure.**

This paper is one of a series dealing with the solubilities of $N_2$, $O_2$, and Ar (*14, 16*), He and Ne (*17*), $^3$He (*15*), and $CO_2$ (*18*) in pure water and seawater. The methods used here for the solubility of krypton follow closely this earlier work. The measurements were made using the same analytical techniques, and the data were fitted to equations in temperature and salinity using the same numerical methods.

This work was prompted by a desire to evaluate solubility relations for krypton concentrations in the sea (*2, 3, 10*). Existing measurements of krypton solubilities were not adequate for this purpose. The only existing measurements of Kr solubility in seawater were by Koenig (*11*), whose data on He, Ne, and Ar solubilities showed low precision and poor agreement with other workers, and by Wood and Caputi (*19*), who made measurements at only three temperatures and one salinity. In distilled water, the measurements of Morrison and Johnstone (*12*) were the most detailed, although they did not agree well with the distilled water data of Koenig (*11*) or Wood and Caputi (*19*).

### Nonideality

At standard conditions, 1 mol of krypton occupies ~0.3% less volume than 1 mol of ideal gas. This difference is about twice the precision of the solubility measurements reported here, so that correction for nonideality is essential. The application of Henry's law to real gases has been reviewed by Weiss (*18*), who showed that the solubility of $CO_2$ over the range 0–40 °C, and at partial pressures up to 500 atm, is well represented by a modified form of Henry's law expressed in terms of the fugacity of the solute and the total pressure. The same approach is followed here for krypton. A more thorough discussion of the pertinent thermodynamics is given in the earlier work (*18*).

The solubility of krypton is given in terms of the Bunsen coefficient $\beta$, defined here as the volume of gas (STP) absorbed per unit volume of the *solution,* at the temperature of the measurement, when the total pressure and the fugacity are both 1 atm. According to the modified form of Henry's law (*18*), $C$, the volume (STP) of gas dissolved per unit volume of *solution* at the temperature of the measurement, is given by the relation

$$C = \beta f \exp[(1 - P)\bar{v}/RT] \qquad (1)$$

where $f$ is the fugacity, $P$ is the *total* pressure, $\bar{v}$ is the partial molal volume of the gas in solution, $R$ is the gas constant, and $T$ is the absolute temperature.

Although $\bar{v}$ for krypton in aqueous solution has not been measured, values of $\bar{v}$ for He, Ar, $N_2$, $O_2$, and $CO_2$ do not differ greatly (*8*). In our calculations, we have used a value of $\bar{v}$ for krypton of 33.5 cm$^3$/mol, which lies between the measured values for Ar and $CO_2$. This value of $\bar{v}$ corresponds to a variation in $C$ of ~0.14% per atmosphere deviation from 1 atm total pressure. Thus, for krypton solubility at *total* pressures near 1 atm, the exponential term in eq 1 may generally be neglected, reducing the equation to the simple form $C = \beta f$.

In the following discussions, gas volumes and fugacity–pressure corrections are based on the virial equation of state. For the pure gas (neglecting the small contribution of water vapor) the solubility measurements have been corrected using the approximations

$$V(P,T) = V^*(P,T) + B(T) \qquad (2)$$

and

$$f = P \exp[B(T)P/RT] \qquad (3)$$

where $V$ is the volume of 1 mol of real gas, $V^*$ is the volume of 1 mol of ideal gas, and $B(T)$ is the second virial coefficient. Values of $B(T)$ in cm$^3$/mol for Kr in the range 270–320 K (*13*) are well represented by the power series

$$B(T) = -516.66 + 3.3769T - (8.3056 \times 10^{-3})T^2 + (7.41404 \times 10^{-6})T^3 \qquad (4)$$

In order to calculate the solubility of atmospheric krypton, the fugacity of krypton in air must be evaluated. Because the mole fraction of krypton in air is $\ll 1$, the atmosphere may be regarded for these purposes as a binary Kr–air mixture. The following equation gives the fugacity of each component in a binary mixture (*18*):

$$f_1 = x_1 P \exp[(B_{11} + 2x_2^2\delta_{12})P/RT] \qquad (5)$$

where the subscripts 1 and 2 refer to the two components of the mixture, $x$ is the mole fraction, $B_{11}$ is the second virial coefficient of pure gas 1, and $\delta_{12}$ equals the cross virial coefficient $B_{12}$ for interactions between the two gases minus the mean of $B_{11}$ and $B_{22}$ for the two pure gases.

Following the treatment used earlier for $CO_2$ in air (*18*), the quantity $\delta_{Kr-air}$ has been evaluated using the Lennard-Jones (6–12) potential model. Over the temperature range −2 to +40 °C, $\delta_{Kr-air}$ may be taken as a constant $+3.5 \pm 0.3$ cm$^3$/mol. The magnitude of this correction, as opposed to assuming $\delta = 0$ (Lewis and Randall rule) is ~0.03%, or about one-eighth of the total deviation from ideal gas behavior. Although we have used the calculated value of $\delta_{Kr-air} = 3.5$ cm$^3$/mol to prepare our fitted equations and tables for the solubility of atmospheric krypton, the 0.03% error introduced by assuming $\delta = 0$ may be tolerable for many applications.

### Experimental Section

Krypton solubilities were measured by the Scholander microgasometric technique as refined by Douglas (*6, 7*) and Weiss (*17, 18*). This method was chosen because of its simplicity, precision, and proven agreement with other high-precision techniques in measuring the solubilities of a number of other gases (*14, 16, 18*). A detailed description of the apparatus and technique is given in the cited literature.

Measurements were made using krypton supplied by Matheson Gas Products and specified >99.995% pure. Independent chromatographic checks showed <0.01% air contamination. Surface seawater, collected at La Jolla and evaporated to increase its salinity by ~2 parts per thousand (‰), was passed through a 0.45 $\mu$m filter and was poisoned with ~1 mg/L of

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**Table I. Experimental Krypton Solubilities: Values of the Bunsen Coefficient $\beta$ at Various Temperatures and Salinities**

| S‰ | t, °C | $10^2 \beta$ | t, °C | $10^2 \beta$ | t, °C | $10^2 \beta$ |
|---|---|---|---|---|---|---|
| 0.0 | 0.59 | 10.778 | 20.24 | 6.222 | 30.98 | 4.955 |
| | 0.57 | 10.778 | 20.24 | 6.220 | 30.97 | 4.961 |
| | 0.57 | 10.790 | 20.25 | 6.227 | 40.09 | 4.215 |
| | 0.58 | 10.786 | 20.24 | 6.221 | 40.10 | 4.218 |
| | 10.33 | 7.981 | 20.25 | 6.226 | 40.10 | 4.216 |
| | 10.32 | 7.985 | 30.97 | 4.953 | | |
| | 10.32 | 7.979 | 30.98 | 4.957 | | |
| 19.046 | 4.90 | 8.200 | 30.08 | 4.525 | 30.08 | 4.532 |
| | 4.90 | 8.212 | 30.08 | 4.510 | | |
| | 4.90 | 8.220 | 30.08 | 4.517 | | |
| 36.595 | 0.09 | 8.451 | 10.42 | 6.276 | 29.96 | 4.084 |
| | 0.07 | 8.438 | 10.41 | 6.270 | 29.97 | 4.093 |
| | 0.09 | 8.442 | 15.10 | 5.566 | 29.96 | 4.085 |
| | 0.08 | 8.444 | 15.10 | 5.547 | 40.10 | 3.475 |
| | 5.32 | 7.198 | 15.10 | 5.554 | 40.10 | 3.469 |
| | 5.31 | 7.204 | 20.34 | 4.920 | 40.10 | 3.481 |
| | 5.32 | 7.193 | 20.34 | 4.933 | 40.09 | 3.475 |
| | 10.42 | 6.273 | 20.34 | 4.935 | | |

$HgCl_2$. Pure water solubilities were measured in distilled water, which was also used to prepare diluted seawater for measurements at lower salinity. An inductive salinometer was used to determine salinities to ±0.004‰.

As in previous solubility measurements by this technique, the equilibration process was found to obey first-order kinetics, with a characteristic time $\tau$ in distilled water ranging from 2.5 min at 40 °C to 11.5 min at 0 °C. Equilibration times in seawater were similar. In all cases the experiment was run for $\sim 10\ \tau$ to ensure complete equilibration.

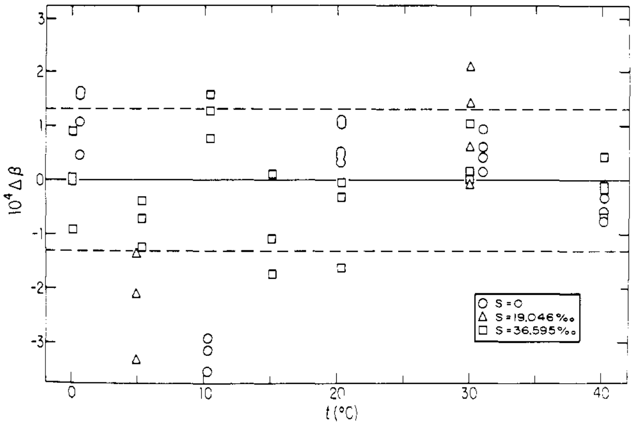

**Figure 1.** Deviations of all the experimental krypton solubilities listed in Table I from the fit to eq 6, plotted against temperature. The dashed lines show ± the root-mean-square deviation.

The results were calculated in the same manner as previous microgasometric measurements, and the corrections for nonideality discussed in the foregoing section were applied. The correction for the expansion of the aqueous phase during equilibration was made using the same estimated partial molal volume used in the nonideality corrections.

## Results

Experimental values of the Bunsen solubility coefficient $\beta$ for krypton at various temperatures and salinities are given in Table I. These data have been corrected for nonideality as discussed above.

**Table II. Constants for Calculations of Krypton Bunsen Solubility Coefficients $\beta$ and Krypton Solubilities in mL/L and mL/kg from Moist Air at 1 Atm Total Pressure According to Eq 6 and 7**

| Units | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $B_1$ | $B_2$ | $B_3$ |
|---|---|---|---|---|---|---|---|
| $\beta$ | −57.2596 | 87.4242 | 22.9332 | | −0.008723 | −0.002793 | 0.0012398 |
| mL/L | −109.9329 | 149.8152 | 72.8393 | −9.9217 | −0.006953 | −0.004085 | 0.0014759 |
| mL/kg | −112.6840 | 153.5817 | 74.4690 | −10.0189 | −0.011213 | −0.001844 | 0.0011201 |

**Table III. Bunsen Coefficients $\times 10^2$ for Krypton**

| | Salinity, ‰ | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| t, °C | 0 | 10 | 20 | 30 | 34 | 35 | 36 | 38 | 40 |
| −1 | | | 9.842 | 9.163 | 8.905 | 8.842 | 8.779 | 8.654 | 8.532 |
| 0 | 10.979 | 10.226 | 9.525 | 8.872 | 8.623 | 8.562 | 8.502 | 8.382 | 8.264 |
| 1 | 10.623 | 9.899 | 9.224 | 8.595 | 8.355 | 8.297 | 8.238 | 8.123 | 8.009 |
| 2 | 10.284 | 9.587 | 8.937 | 8.331 | 8.100 | 8.043 | 7.987 | 7.876 | 7.766 |
| 3 | 9.962 | 9.290 | 8.664 | 8.079 | 7.857 | 7.802 | 7.748 | 7.640 | 7.535 |
| 4 | 9.654 | 9.007 | 8.403 | 7.840 | 7.625 | 7.572 | 7.520 | 7.416 | 7.314 |
| 5 | 9.362 | 8.737 | 8.155 | 7.611 | 7.404 | 7.353 | 7.303 | 7.202 | 7.104 |
| 6 | 9.082 | 8.480 | 7.918 | 7.393 | 7.193 | 7.144 | 7.095 | 6.998 | 6.903 |
| 8 | 8.561 | 8.000 | 7.476 | 6.987 | 6.800 | 6.754 | 6.708 | 6.618 | 6.529 |
| 10 | 8.086 | 7.563 | 7.073 | 6.616 | 6.441 | 6.398 | 6.355 | 6.271 | 6.187 |
| 12 | 7.653 | 7.163 | 6.705 | 6.277 | 6.113 | 6.073 | 6.033 | 5.954 | 5.875 |
| 14 | 7.256 | 6.798 | 6.369 | 5.967 | 5.813 | 5.775 | 5.738 | 5.663 | 5.590 |
| 16 | 6.892 | 6.463 | 6.060 | 5.682 | 5.538 | 5.503 | 5.467 | 5.397 | 5.328 |
| 18 | 6.558 | 6.155 | 5.777 | 5.422 | 5.286 | 5.252 | 5.219 | 5.153 | 5.088 |
| 20 | 6.252 | 5.873 | 5.517 | 5.182 | 5.054 | 5.023 | 4.991 | 4.929 | 4.868 |
| 22 | 5.970 | 5.613 | 5.277 | 4.962 | 4.841 | 4.811 | 4.782 | 4.723 | 4.665 |
| 24 | 5.710 | 5.374 | 5.057 | 4.759 | 4.645 | 4.616 | 4.588 | 4.533 | 4.478 |
| 26 | 5.471 | 5.153 | 4.854 | 4.572 | 4.464 | 4.437 | 4.411 | 4.358 | 4.306 |
| 28 | 5.249 | 4.949 | 4.666 | 4.399 | 4.297 | 4.272 | 4.246 | 4.197 | 4.148 |
| 30 | 5.045 | 4.761 | 4.493 | 4.240 | 4.143 | 4.119 | 4.095 | 4.048 | 4.001 |
| 32 | 4.856 | 4.587 | 4.333 | 4.092 | 4.000 | 3.977 | 3.955 | 3.910 | 3.866 |
| 34 | 4.681 | 4.426 | 4.184 | 3.956 | 3.869 | 3.847 | 3.825 | 3.783 | 3.741 |
| 36 | 4.518 | 4.276 | 4.047 | 3.830 | 3.747 | 3.726 | 3.706 | 3.665 | 3.625 |
| 38 | 4.368 | 4.138 | 3.920 | 3.713 | 3.634 | 3.614 | 3.595 | 3.556 | 3.518 |
| 40 | 4.228 | 4.009 | 3.802 | 3.605 | 3.530 | 3.511 | 3.492 | 3.455 | 3.419 |

Table IV. Solubility of Krypton from Moist Air at 1 Atm Total Pressure, in Units of $10^{-5}$ mL/kg

| $t$, °C | Salinity, ‰ | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | 0 | 10 | 20 | 30 | 34 | 35 | 36 | 38 | 40 |
| −1 | | | 10.953 | 10.118 | 9.802 | 9.724 | 9.647 | 9.495 | 9.346 |
| 0 | 12.412 | 11.470 | 10.600 | 9.795 | 9.491 | 9.416 | 9.342 | 9.196 | 9.052 |
| 1 | 12.008 | 11.101 | 10.263 | 9.488 | 9.195 | 9.123 | 9.052 | 8.911 | 8.772 |
| 2 | 11.622 | 10.749 | 9.942 | 9.195 | 8.912 | 8.843 | 8.774 | 8.638 | 8.505 |
| 3 | 11.254 | 10.413 | 9.636 | 8.916 | 8.643 | 8.576 | 8.510 | 8.379 | 8.250 |
| 4 | 10.903 | 10.093 | 9.343 | 8.649 | 8.386 | 8.322 | 8.258 | 8.131 | 8.006 |
| 5 | 10.568 | 9.787 | 9.064 | 8.394 | 8.141 | 8.078 | 8.017 | 7.894 | 7.774 |
| 6 | 10.248 | 9.495 | 8.797 | 8.151 | 7.906 | 7.846 | 7.786 | 7.668 | 7.552 |
| 8 | 9.650 | 8.949 | 8.299 | 7.696 | 7.467 | 7.411 | 7.355 | 7.245 | 7.137 |
| 10 | 9.103 | 8.449 | 7.843 | 7.279 | 7.065 | 7.013 | 6.961 | 6.858 | 6.756 |
| 12 | 8.602 | 7.991 | 7.424 | 6.897 | 6.697 | 6.648 | 6.599 | 6.502 | 6.407 |
| 14 | 8.141 | 7.570 | 7.039 | 6.546 | 6.358 | 6.312 | 6.266 | 6.176 | 6.086 |
| 16 | 7.718 | 7.183 | 6.685 | 6.222 | 6.046 | 6.003 | 5.960 | 5.875 | 5.791 |
| 18 | 7.327 | 6.826 | 6.359 | 5.924 | 5.758 | 5.718 | 5.677 | 5.597 | 5.519 |
| 20 | 6.967 | 6.496 | 6.057 | 5.648 | 5.492 | 5.454 | 5.416 | 5.341 | 5.267 |
| 22 | 6.633 | 6.191 | 5.778 | 5.393 | 5.246 | 5.210 | 5.175 | 5.104 | 5.034 |
| 24 | 6.324 | 5.908 | 5.520 | 5.157 | 5.018 | 4.984 | 4.951 | 4.884 | 4.818 |
| 26 | 6.038 | 5.646 | 5.280 | 4.937 | 4.807 | 4.775 | 4.743 | 4.680 | 4.617 |
| 28 | 5.771 | 5.402 | 5.057 | 4.733 | 4.610 | 4.580 | 4.549 | 4.490 | 4.431 |
| 30 | 5.523 | 5.175 | 4.849 | 4.544 | 4.427 | 4.398 | 4.369 | 4.313 | 4.257 |
| 32 | 5.292 | 4.964 | 4.656 | 4.366 | 4.256 | 4.229 | 4.202 | 4.148 | 4.095 |
| 34 | 5.077 | 4.766 | 4.475 | 4.201 | 4.096 | 4.071 | 4.045 | 3.994 | 3.944 |
| 36 | 4.875 | 4.581 | 4.306 | 4.047 | 3.947 | 3.923 | 3.899 | 3.850 | 3.803 |
| 38 | 4.686 | 4.409 | 4.147 | 3.902 | 3.808 | 3.784 | 3.761 | 3.716 | 3.671 |
| 40 | 4.509 | 4.247 | 3.999 | 3.766 | 3.677 | 3.655 | 3.633 | 3.589 | 3.547 |

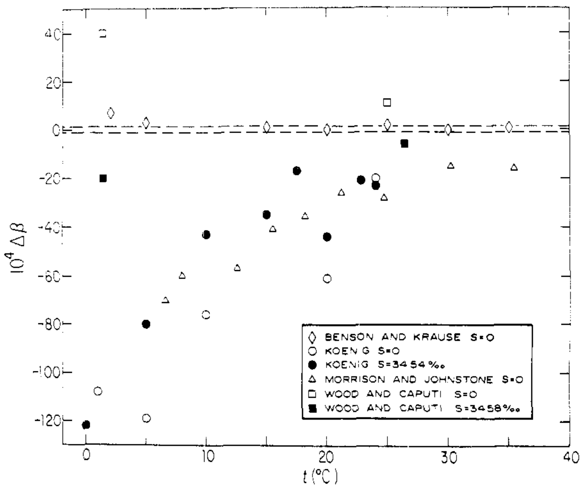

**Figure 2.** Deviations of krypton solubility data from other laboratories from the present work as given by the fit to eq 6, plotted against temperature. The dashed lines represent ± the root-mean-square deviation of the present experimental data from the fitted curve (compare vertical scales with the same lines shown in Figure 1).

Equations in temperature and salinity which give the Bunsen solubility coefficient, as well as the solubility from moist air at 1 atm total pressure in units of milliliters (STP)/liter and milliliters (STP)/kilogram have been used to fit the krypton solubility data reported here. These equations make use of the integrated van't Hoff equation and the salinity dependence of the Setchénow relation, which prescribes a linear relationship between salt concentration and the logarithm of the solubility. Previous work has shown that these equations represent accurately the solubilities of nitrogen, oxygen, argon, helium, neon, carbon dioxide, methane, and hydrogen in distilled water and seawater (*5, 14, 16, 17, 18, 20*). For krypton, as was the case for the other gases, a three-term expansion of the temperature dependence of the Bunsen coefficient was found to fit the data to within the precision of the measurement. For detailed discussions of the equations, their derivation, and the data fitting procedures, the reader is referred to the original paper (*14*).

The Bunsen solubility coefficient $\beta$ of krypton, corrected for nonideal behavior, is given by an equation of the form

$$\ln \beta = A_1 + A_2(100/T) + A_3 \ln (T/100) + S‰ [B_1 + B_2(T/100) + B_3(T/100)^2] \quad (6)$$

where the $A$'s and $B$'s are constants, $T$ is the absolute temperature, and $S‰$ is the salinity in parts per thousand. In fitting the data to this equation the points measured in diluted seawater (salinity 19.046‰) were omitted from the fitting procedure and were used to check the validity of the Setchénow salting-out relationship. As was the case with the other gases, the data support the use of the Setchénow treatment.

Solubilities from moist air in milliliters/liter and milliliters/kilogram are given by an equation of the form

$$\ln C^* = A_1 + A_2(100/T) + A_3 \ln (T/100) + A_4(T/100) + S‰ [B_1 + B_2(T/100) + B_3(T/100)^2] \quad (7)$$

in which $C^*$ is the atmospheric solubility in milliliters(STP)/liter (at the temperature of the measurement), or in milliliters (STP)/kilogram, from water-saturated air at a total pressure of 1 atm. The equation is similar in form to eq 6, except that an additional term in temperature is required (*14*). In these calculations, the mole fraction of krypton in dry air was taken as $1.141 \times 10^{-6}$. This value was calculated from the volume fraction data of Glueckauf and Kitt (*9*) corrected for nonideality. Similarly, all the data fitted to eq 7 were first corrected for nonideality.

Constants obtained by fitting the experimental data to eq 6 and 7 for values of $\beta$ and $C^*$ in milliliters/liter and milliliters/kilogram are given in Table II. Figure 1 shows the deviations of the measured data from the fit to eq 6. The root-mean-square deviation of all the data from the fitted curve is $1.3 \times 10^{-4}$ in units of $\beta$, or about 0.2%. The solubility of krypton in units of $\beta$ and milliliters/kilogram at various temperatures and salinities is given in Tables III and IV.

In Figure 2, the results of previous studies of krypton solubility (*11, 12, 19*) are compared to the present measurements. Also plotted are the recently published data for the solubility of krypton in distilled water from the concurrent study of Benson and Krause (*1*). Note that the vertical scale in Figure 2 is considerably less sensitive than in Figure 1, as is shown by the dashed lines representing the root-mean-square deviation of the present data which are plotted in both figures. The data of Koenig (*11*) show the greatest deviations from the present work, especially at lower temperatures where his values are as much as 14% below ours. The Morrison and Johnstone data (*12*) also lie below our values, by up to 8% at the lower temperatures. The data of Wood and Caputi (*19*) are in better agreement: their two distilled water points which lie within our temperature range average 2.9% above our values, and their two seawater points within our temperature range average 1.9% below our values. However, the comparison of our data with those of Benson and Krause (*1*) is especially striking. Their seven distilled water measurements within our temperature range average just 0.24% above our values, with a maximum difference of 0.6%. Thus, although our methods give slightly different values for He and Ne solubilities (*1*), the two independent techniques show remarkably similar results for the solubility of krypton in distilled water.

### Conclusions

The data reported here greatly improve our knowledge of the solubility of krypton, especially in seawater. The fitted values have an estimated accuracy of $\pm$0.4% and a relative precision ($\pm$one standard deviation) of about 0.2%. The reliability of these results is strengthened by the accuracy of the solubilities of other gases measured by the same technique and by the close agreement with the recent data of Benson and Krause for the solubility of krypton in distilled water.

We have made a preliminary investigation of saturation anomalies shown by various measurements of krypton dissolved in the sea (*2, 3, 10*). Following the approach used earlier to study the relative enrichments of He, Ne, and Ar (*4*), an analysis of Ne, Ar, and Kr saturation anomalies shows that Kr fits a consistent pattern of decreasing enrichment with increasing $\beta$, which may be explained by the injection of $\sim$1 mL of air/kg of seawater in excess of equilibrium solubility. A detailed evaluation of these effects is reserved for later discussion.

### Acknowledgment

We thank H. Craig and S. L. Miller for their interest, encouragement, and advice.

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Received for review July 13, 1977. Accepted October 21, 1977. This work was supported by a U.S. National Science Foundation grant to the Isotope Laboratory, Scripps Institution of Oceanography.

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# Additivity of Volumes in Hydrated Melts: Mixtures of Calcium and Cadmium Nitrate Tetrahydrates with Ammonium Nitrate

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**Densities of $Ca(NO_3)_2 \cdot 4H_2O + NH_4NO_3$ and $Cd(NO_3)_2 \cdot 4H_2O + NH_4NO_3$ systems have been measured at temperatures between 285 and 365 K and up to 60 equiv % of $NH_4NO_3$. The systems obey the principle of additivity of volumes. Densities and equivalent volumes vary linearly with temperature.**

It has been realized (*1–4, 7, 9–17*) that the studies of highly concentrated aqueous electrolyte solutions and hydrated melts are of importance in correlating the behavior of anhydrous molten salts and dilute aqueous electrolyte solutions. Recently, there has been special interest in the use of hydrated melts and highly concentrated aqueous electrolyte solutions, with supercooling and glass-forming tendencies, in certain specialized fields of research. The principle of additivity of volumes has been found to be a useful approximation (*2, 10*) in evaluating the volumes at any desired composition from the limited data. The present investigation was aimed at exploring the validity of this approximation for $(Ca,Cd)(NO_3)_2 \cdot 4H_2O + NH_4NO_3$ mixtures.

### Experimental Section

Hydrated calcium and cadmium nitrates were AnalaR (BDH) grade. Water content of the salts, determined by volumetric titration with EDTA, was found to be 4 $\pm$ 0.01 mol/mol of cation. Ammonium nitrate, also AnalaR (BDH), was vacuum desiccated for 2–3 days and stored over anhydrous magnesium perchlorate until used. Mixtures of varying composition were prepared individually by fusing the calculated amounts of the components in a stoppered glass vessel and aging at 50–60 °C for about 4 h before use.

The manometric densitometer (*8*) was modified (*10, 16*) to allow direct measurement of the volume of a known amount of the melt with a precision of $\pm$0.005 $cm^3$. Details regarding calibration of densitometer, dilation correction, thermostat bath, and temperature control unit have been described earlier (*16*). Data were taken both in heating and cooling cycles, and temperature was maintained with a precision of $\pm$0.05 °C.

### Results and Discussion

The density and equivalent volume data for the different compositions of $(Ca,Cd)(NO_3)_2 \cdot 4H_2O + NH_4NO_3$ mixtures con-

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