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*An Experimental Study of the P-V-T-S Relations of Seawater*¹

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ABSTRACT

The compressibility of seawater has been determined as a function of salinity, pressure, and temperature. Specific volumes have been computed. The salinities, as measured on a University of Washington-type conductivity bridge, were 0.00, 30.52, 34.99, and 41.03‰. The pressure range was from 1 to 1300 bars and the temperature range from 0°C to 25°C. The experiments were carried out by measuring decrements of volume at 100-bar intervals spaced in 5° isotherms. The apparatus consisted of a piston-and-cylinder volumometer for measuring volume changes, supplemented by a pycnometer to determine the one-atmosphere specific volumes. The precision of measurements was 7 parts in 10⁵.

The experimental results for distilled water ($S = 0\text{‰}$) are intermediate between five sets of measurements by previous investigators. The pycnometric measurements served to confirm the currently accepted values of the specific volumes of seawater at one bar total pressure as a function of temperature and salinity. The compressibilities of the seawater, however, indicate a systematic density difference of up to 5 parts in 10⁴ at the highest pressures between our measurements and currently accepted values, our values trending lower. Such a difference affects oceanographic calculations as based on currently accepted values of specific volume.

Introduction. Precise P-V-T data for water have always been in great demand in both engineering and scientific endeavors. These demands are currently met by the work of only a few investigators, notably E. H. Amagat (1893), Smith and Keyes (1934), Kennedy et al. (1958), and Jůza (1963), mostly in noncoinciding ranges of pressure and temperature. Where the data do coincide, it is of greatest importance to compare them as the comparison provides an idea of the precision on which reliance can be placed in making thermodynamic calculations. Eckart (1958b) has pointed out that the several sets of data in the lower temperature range (0°C to 50°C) for pure water contain random errors of up to 2 parts in 10⁴ at pressures near 1000 bars and that they also differ systematically. The systematic differences, of

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course, emphasize the need for data from different types of experimental apparatus.

The existing P-V-T data for seawater have been obtained in the same manner as those for pure water or other fluids. In addition to furnishing the basis for a general equation to represent the physical state of the oceans under different conditions of depth, temperature, and salinity, these data are used directly in certain oceanographic calculations, such as those of dynamic height and geostrophic velocity. These quantities are related to the motion of currents consequent on the ocean's density field. Moreover, derived thermodynamic functions, the compressibility and thermal expansion, are used to calculate sound velocities and adiabatic temperature gradients in the oceans.

The P-V-T data required for making these calculations are needed to a precision of 1 part in 10⁵. It is remarkable that the *currently accepted* values of the specific volume of seawater as a function of pressure, temperature, and salinity stem from only one set of measurements. Knudsen (1901) measured some 1-atmosphere specific volumes at differing temperatures and salinities, and Ekman (1908) measured compressibility of seawater of two different salinities at three temperatures and three pressures. Although the E-K values are remarkably consistent as regards random errors (Eckart 1958b), they are not likely to be more precise than values for pure water, and may contain systematic errors. Crease (1962) calculated specific volume from recently acquired velocity data (Wilson 1960). The purpose of our study was to provide additional P-V-T data for seawater in the range of natural conditions by using a very different experimental method so that an effective comparison could be made with the E-K values.

Though agreement is practically general that the E-K specific-volume tables are reliable to 1 or 2 parts in 10⁴, Eckart (1958b) has stated that their current tabulation and use to six or more decimal places is misleading. Reid (1958), who seems to be satisfied that the present compressibility data are accurate enough for computing geostrophic velocities, has pointed out that thermal expansion is the quantity that must be accurately known. Eckart (1958a) has also stated that isopiestic thermal expansions at higher pressures suffer the same limitations as the compressibility data. Fofonoff (1963) and Wilson (1958) have shown that the present equation of state for seawater is not nearly of sufficient precision for sound-velocity calculations to the nearest 0.2 m/sec. Eckart (1958a) stated further that the equation of state is not precise enough to determine the stability of the deep ocean layers.

Experimental Apparatus. High-pressure volumetry has figured in a number of studies in our laboratory, and much of the apparatus has been described elsewhere (Kennedy 1954, Kennedy et al. 1958, Sourirajan and Kennedy 1962, 1963). The present study, however, utilized several new components especially designed for this work, as well as an entirely different operational

procedure. The experimental system consisted of a volumometer of approximately 20 cc in which the compressibility measurements were made, another volumometer of 6 cc that served to correct for the compressibility of auxiliary portions of the system, a bellows-type fluid separator, and Aminco steel valve blocks with high-pressure capillary tubing. Pressures were measured with a 1400-bar Heise Bourdon-type pressure gauge; our gauge was standardized against a dead-weight piston. Temperatures were controlled by means of an open box-like copper water bath that enclosed the larger volumometer. Temperatures were measured with a Braun 75-mm thermometer calibrated in 10ths of a degree from 0°C to 100°C ; this thermometer registered 100°C in boiling water, 0°C in an equilibrium mixture of ice and water, and agreed with a U.S. Bureau of Standards calibrated thermocouple to within the reading accuracy. Since the conditions of this study ranged from 0°C to 25°C , temperatures were controlled by the addition at regular intervals of either a little ice or a little hot water to the constant-temperature bath. In this way it was easy to control the temperature to within 0.1°C . Samples of water under study were stored in a 100-ml burette permanently connected to the experimental system by means of small-bore surgical tubing, safety-wired at both ends and heavily coated with stopcock grease. Approximately one inch of clear, heavy mineral oil (Nujol) was floated on the reservoir surface of the sample fluid, completely preventing evaporation. Fig. 1 is a schematic representation of the apparatus.

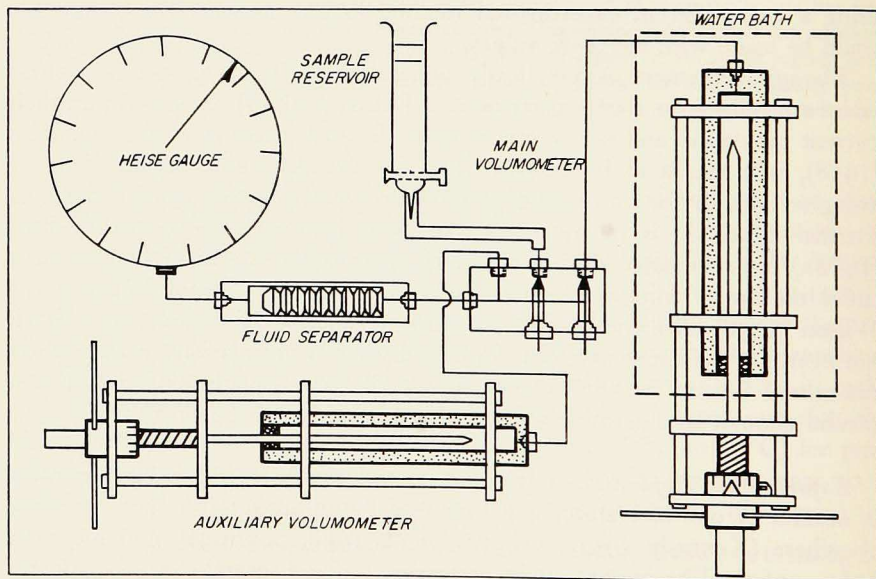


Figure 1. Diagram of the experimental apparatus.

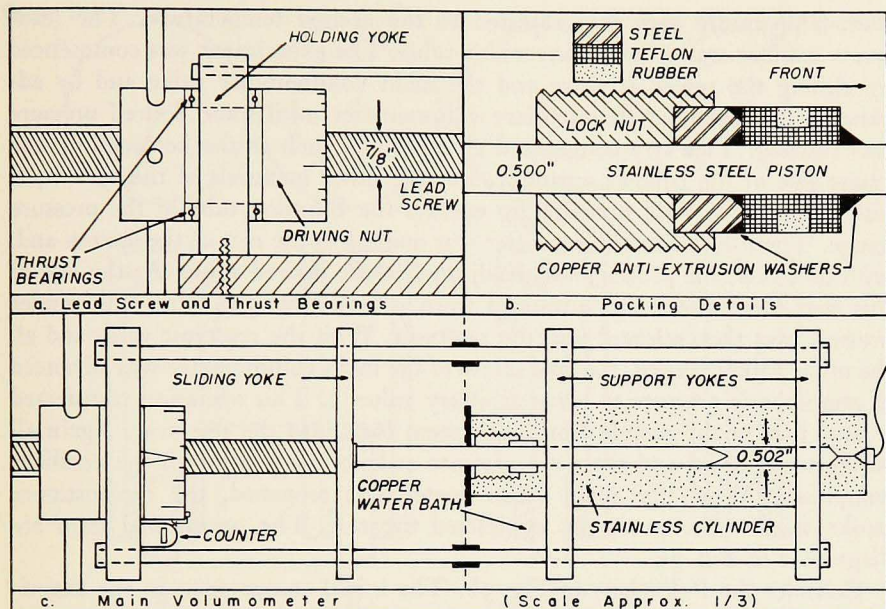


Figure 2. Main volumometer.

The volumometer in which the compressibility measurements were carried out consisted of a screw-driven piston and stainless-steel cylinder anchored with heavy yokes and columns; the cylinder was 13 inches (33 cm) long and 3 inches (7.6 cm) in diameter. A central cylindrical chamber, 0.502 inches (1.275 cm) in diameter, was drilled the length of the cylinder and was closed at the front end by an Aminco pressure fitting and capillary line. Fluids were compressed in the cylindrical chamber with a stainless-steel piston, 0.500 inches (1.270 cm) in diameter. The total stroke, 7 inches (17.8 cm), was designed to void the cylinder when fully inserted. The piston was carried and supported by a sliding yoke driven by a 16-inch (40.6-cm), Acme thread lead screw, $7/8$ inches (2.22 cm) in diameter with a pitch of 10. Lead screw and piston were advanced by a driving nut calibrated to 0.01 turn. The lead screw, manufactured and precision-ground by the Waltham Machine Works, was found to displace 0.3190 cc per turn, to a precision of 1 part in 10⁵ throughout its entire stroke (Kennedy 1954). Unsupported area packings of rubber and teflon with a locknut were used to seal the rear end of the volumometer. Details of the volumometer and packings are shown in Fig. 2.

Experimental Procedure. The system was filled with the sample fluid by opening the valve of the reservoir and the valves of both volumometers, and bringing both pistons to the rear of their stroke. At the same time, the con-

stant-temperature bath was adjusted to the desired temperature. The lead-screw counter was set at a convenient value. The experiment was commenced by closing the reservoir valve and the main volumometer valve and by advancing the piston of the auxiliary volumometer until some desired pressure was reached. This step compressed all the fluid, such as the kerosene on the gauge side of the bellows separator, and the other materials of the system; it also supplied the fluid necessary to operate the Bourdon tube of the pressure gauge. Then the main volumometer was opened to the rest of the system and, with its screw, the pressure was readjusted to the desired value. At the end of this step the counter had advanced from a to b (Fig. 3a). The fluid under pressure was then released into the reservoir. With the reservoir valve and all the other valves opened, the lead screw of the main volumometer was advanced at atmospheric pressure to some arbitrary value, c . This operation transferred a large part of the sample from the system back into the reservoir. Again all valves were closed and the same pressure as before was set with the auxiliary compressor. When the main volumometer was reopened, the readjustment stroke was from c to d ; this completed the run. The operational steps are diagramed in Fig. 3a.

Consider the P-V plane in Fig. 3b. The initial compression in the experiment, from a to b , was carried out with a mass of fluid, m_1 , in the volumometer. At b , the pressure was p , and the volume may be designated V_1 . The second compression, from c to d , brought a smaller mass of fluid, m_2 , to the same pressure, p , where its volume was V_2 . If the specific volume of the fluid at that pressure was \tilde{V}_p , then

$$\tilde{V}_p = \frac{V_1}{m_1} = \frac{V_2}{m_2}, \quad (1)$$

and $\tilde{V}_p m_1 = V_1$, while $\tilde{V}_p m_2 = V_2$. Subtracting these two equalities yields

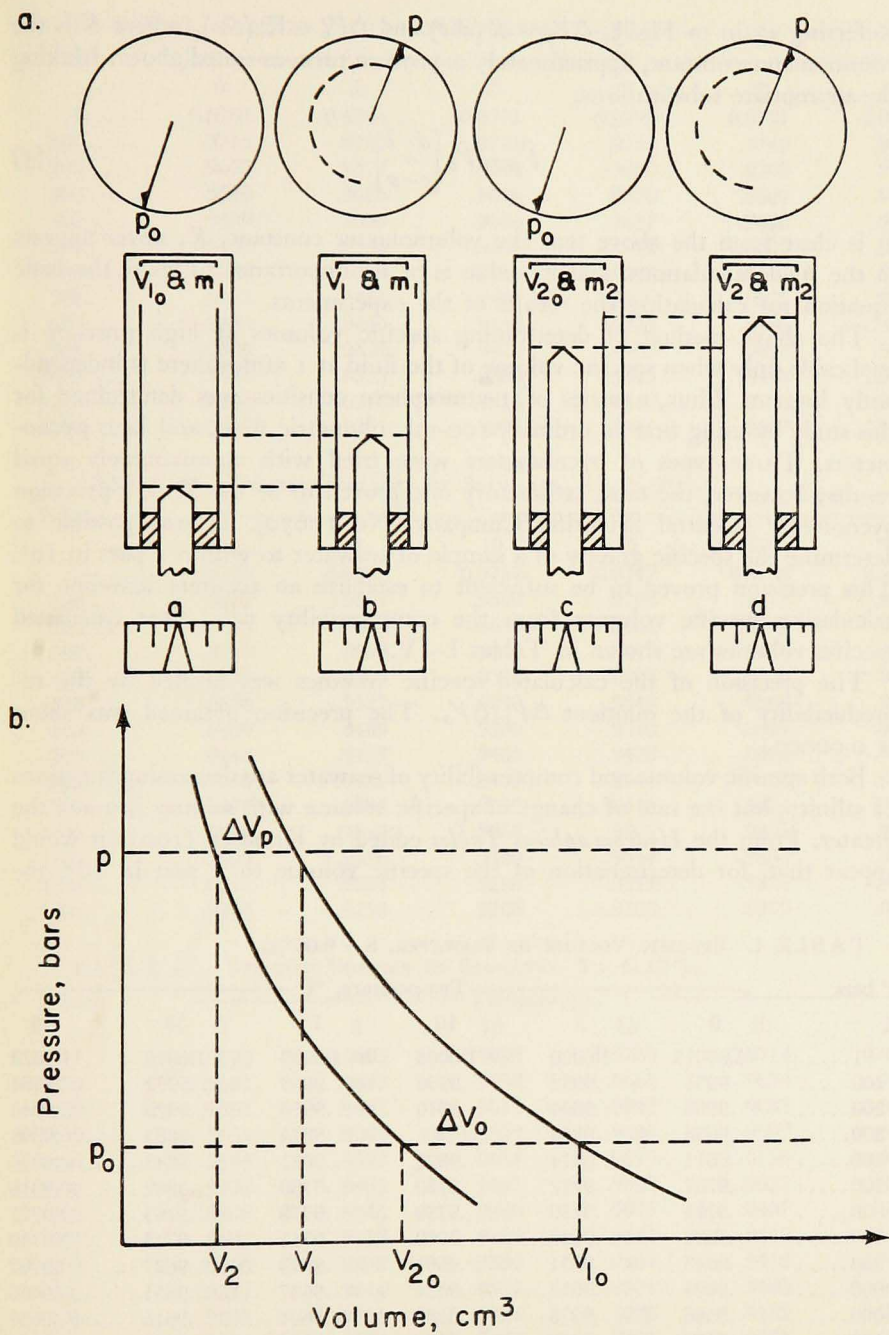
$$\tilde{V}_p = \frac{\Delta V_p}{\Delta m}. \quad (2)$$

The quantity m can be determined experimentally by gathering and weighing the fluid ejected in the stroke from b to c ; however, this procedure is inaccurate with the present apparatus. Instead,

$$\tilde{V}_o = \frac{V_{1o}}{m_1} = \frac{V_{2o}}{m_2}, \quad (3)$$

where the subscripts o refer to the volumes at atmospheric pressure. Therefore $\tilde{V}_o = \Delta V_o / \Delta m$, and

$$\tilde{V}_p = V_o \left[\frac{\Delta V_p}{\Delta V_o} \right]. \quad (4)$$



Referring again to Fig. 3, $\Delta V_p = K(d-b)$ and $\Delta V_o = K(c-a)$, where K is the volumometer constant, approximately 0.3190 cc/turn, as noted above. Making the appropriate substitutions,

$$\tilde{V}_p = \tilde{V}_o \left[\frac{d-b}{c-a} \right]. \quad (5)$$

It is clear from the above that the volumometer constant, K , never appears in the final calculations, and its value is of no importance. This is the basic equation for calculating the results of the experiments.

The above method of determining specific volumes at high pressure is applicable only when specific volume of the fluid at 1 atmosphere is independently known. Thus, a series of 1-atmosphere densities was determined for this study by using first an ordinary 100-ml volumetric flask, and later pycnometers. Three types of pycnometers were tried with approximately equal results; however, the most satisfactory one proved to be the Weld's precision pycnometer (Central Scientific Company, No. 10670). It was possible to determine the specific gravity of a sample of seawater to within 1 part in 10^5 . This precision proved to be sufficient to establish an accurate base-line for calculating specific volumes from the compressibility data. Our calculated specific volumes are shown in Tables I-IV.

The precision of the calculated specific volumes was limited by the reproducibility of the quotient $\Delta V_p/\Delta V_o$. The precision obtained was about ± 0.00007 .

Both specific volume and compressibility of seawater are decreasing functions of salinity, but the rate of change of specific volume with salinity is much the greater. From the *Hydrographical Tables* edited by Knudsen (1901) it would appear that, for determination of the specific volume to 1 part in 10^4 , the

TABLE I. SPECIFIC VOLUME OF SEAWATER. $S = 0.00\text{‰}$.

P bars	Temperature, °C					
	0	5	10	15	20	25
1.....	1.00013	1.0000	1.0003	1.0009	1.0018	1.0029
100.....	.9951	.9952	.9956	.9963	.9972	.9984
200.....	.9903	.9904	.9910	.9918	.9928	.9940
300.....	.9856	.9858	.9865	.9874	.9885	.9898
400.....	.9811	.9814	.9822	.9831	.9843	.9856
500.....	.9767	.9772	.9780	.9790	.9802	.9816
600.....	.9724	.9730	.9739	.9750	.9763	.9777
700.....	.9683	.9690	.9700	.9711	.9725	.9739
800.....	.9643	.9651	.9661	.9673	.9687	.9702
900.....	.9604	.9613	.9624	.9637	.9651	.9666
1000.....	.9566	.9576	.9587	.9601	.9615	.9631
1100.....	.9529	.9540	.9552	.9566	.9581	.9598
1200.....	.9493	.9505	.9517	.9532	.9547	.9565
1300.....	.9459	.9471	.9484	.9499	.9515	.9533

TABLE II. SPECIFIC VOLUME OF SEAWATER. $S = 30.52\text{‰}$.

P bars	Temperature, °C					
	0	5	10	15	20	25
1.....	0.9761	0.9764	0.9771	0.9779	0.9791	0.9804
100.....	.9715	.9720	.9728	.9737	.9749	.9762
200.....	.9672	.9677	.9686	.9696	.9708	.9722
300.....	.9629	.9636	.9646	.9656	.9669	.9683
400.....	.9589	.9596	.9606	.9617	.9630	.9645
500.....	.9547	.9556	.9567	.9579	.9592	.9608
600.....	.9508	.9518	.9530	.9542	.9556	.9572
700.....	.9471	.9481	.9493	.9506	.9521	.9537
800.....	.9434	.9445	.9458	.9471	.9486	.9502
900.....	.9398	.9409	.9423	.9437	.9452	.9469
1000.....	.9364	.9375	.9389	.9403	.9418	.9435
1200.....	.9298	.9311	.9325	.9340	.9356	.9374
1300.....	.9267	.9281	.9295	.9311	.9327	.9345

TABLE III. SPECIFIC VOLUME OF SEAWATER. $S = 34.99\text{‰}$.

P bars	Temperature, °C					
	0	5	10	15	20	25
1.....	0.9726	0.9730	0.9737	0.9747	0.9758	0.9772
100.....	.9682	.9687	.9695	.9705	.9717	.9731
200.....	.9639	.9645	.9654	.9664	.9677	.9691
300.....	.9598	.9605	.9614	.9625	.9638	.9652
400.....	.9557	.9566	.9575	.9587	.9600	.9615
500.....	.9518	.9527	.9537	.9549	.9563	.9578
600.....	.9480	.9489	.9500	.9513	.9527	.9542
700.....	.9443	.9453	.9465	.9478	.9492	.9508
800.....	.9406	.9417	.9430	.9444	.9458	.9474
900.....	.9371	.9383	.9396	.9410	.9424	.9441
1000.....	.9337	.9349	.9363	.9377	.9392	.9409
1100.....	.9303	.9316	.9330	.9344	.9360	.9377
1200.....	.9271	.9284	.9298	.9313	.9329	.9347
1300.....	.9240	.9253	.9268	.9283	.9299	.9317

TABLE IV. SPECIFIC VOLUME OF SEAWATER. $S = 41.03\text{‰}$.

P bars	Temperature, °C					
	0	5	10	15	20	25
1.....	0.9680	0.9685	0.9693	0.9703	0.9714	0.9728
100.....	.9637	.9643	.9652	.9662	.9674	.9688
200.....	.9595	.9602	.9611	.9622	.9635	.9649
300.....	.9554	.9562	.9572	.9583	.9597	.9611
400.....	.9514	.9523	.9534	.9545	.9559	.9574
500.....	.9476	.9485	.9497	.9509	.9523	.9538
600.....	.9438	.9448	.9460	.9473	.9487	.9502
700.....	.9402	.9413	.9425	.9438	.9453	.9468
800.....	.9366	.9378	.9390	.9404	.9419	.9435
900.....	.9332	.9344	.9357	.9371	.9386	.9402
1000.....	.9298	.9311	.9324	.9338	.9353	.9370
1100.....	.9265	.9278	.9292	.9307	.9322	.9339
1200.....	.9233	.9247	.9261	.9276	.9292	.9309
1300.....	.9202	.9216	.9231	.9246	.9262	.9279

salinity must be known to the nearest part per 5000. Preliminary attempts were made in this laboratory to determine the salinity of seawater samples by precipitating and weighing salts from a sample of known weight. This method, although it gave satisfactory results for the purpose, was later abandoned in favor of more standard methods. All the salinities reported in this work were determined with the University of Washington-type conductivity-bridge apparatus and the Knudsen titration apparatus, at the Marine Physical Laboratory, Point Loma, San Diego, California. Both methods are capable of much more accuracy than were needed here (Paquette 1958). A sample of standard seawater of salinity $S = 34.99$ ppt (so-called Copenhagen seawater) was reported as 35.01‰ on the Point Loma bridge—an apparent accuracy of 2 parts in 105.

All seawater samples used in this study with the exception of the bottled samples of Copenhagen water were taken from the pier of the Scripps Institution of Oceanography, La Jolla, California. The water was first cleared of suspended inorganic and biotic matter by millipore filtration and then stored in five-gallon carboys. Ocean water at La Jolla averages about 34.2‰ , and samples of salinities greater than this were prepared at La Jolla by slow evaporation. Lower salinities were prepared by dilution following shipment of the samples to Los Angeles. About one drop of chloroform per gallon of seawater was added to the samples before storing to prevent growth of algae and diatoms.

Formulation and Interpretation of Results. Various sources of error must be taken into account in order to calculate the most accurate possible specific volumes. Since the volume of the system outside of the main cylinder remains constant at a given pressure, error arising from the compressibility of the fluids and from mechanical components in this part of the system are eliminated by the auxiliary cylinder. Fluids required to operate the bellows of the separator and the Bourdon tube of the Heise gauge are supplied independently of the main cylinder. Errors arising from the pressure expansion and thermal expansion of the main cylinder are eliminated by the subtraction ΔV_p because they have the same values at a given pressure with any volume in the main cylinder.

Isotropic compression of the main piston requires a correction that will have a different value in each leg of the experiment because of the two different volumes of piston in the cylinder. Therefore it was necessary to recalculate the specific volume to take this correction into account.

Fortunately, the compressibility of stainless steel is well known; the correction for the compressibility of the stainless-steel piston amounts to approximately 8 parts in 104 at 1300 bars pressure. This calculation, along with other details of a technical nature, are available in M.S. Newton's unpublished Masters thesis (on file in the Geology Library at the University of California, Los Angeles, # LD 791.8. G3N485).

Another source of error is the air introduced into the system each time it is opened for repairs or for changing samples. The procedure for getting rid of the gas is very time-consuming and wasteful of sample. The apparatus must be pressurized over a period of at least 24 hours, and then the gas-charged sample must be voided; sometimes the process must be repeated several times until satisfactory measurements are possible. However, the error introduced by use of an air-saturated sample is almost certainly not great, for it must be remembered that our method involves measurement of the difference in compressibilities of various samples and not of differences in specific volume. Our densities were always determined by multiplying an experimental compressibility by an independently determined, one-atmosphere specific volume. Thus seawater samples with temperature or chemical composition sufficiently different to give specific volume or density differences of several parts in 10^4 will have essentially identical compressibilities.

To our surprise, conductivity measurements on seawater extracted from the apparatus after contact with the stainless steel always showed a salinity decrease of approximately 0.3‰ . This salinity change would throw all our results in doubt providing we were directly measuring density at high pressures. However, since we are measuring compressibilities, our results are not affected even in the decimal place beyond the figures reported. The compressibilities of samples of seawater, differing in salinity by 0.3‰ , are identical to the limits of our precision.

Similarly, the compressibilities of samples separated in temperature by a few tenths of a degree are essentially identical. Thus, precision of thermometer calibration is not of importance, but it is important to know that temperature is held constant during a compressibility measurement.

All the uncertainties of temperature, changing salinity in the apparatus, and air contamination are small compared to the limit of the precision of the compressibility measurement; this limit or uncertainty affected our final result by approximately 7 parts in 10^5 .

Comparison of our results with those of other investigators is provided in Figs. 4, 5, and 6. The abscissae are pressure in bars; the ordinates are residuals of specific volume—in this case, the values of specific volume of the other investigators minus those of our work times 10^4 . Fig. 4 illustrates the scatter in the existing data for distilled water; it is worth re-emphasizing that the data are much more abundant than for seawater. At 0°C , the results of our study agree, to within the limit of experimental error, with those of Amagat (1893), Ekman (1908), and Crease (1962). In particular, the agreement with Amagat is perfect; these values are intermediate between the Ekman and Crease data—perhaps a favorable indication of accuracy. The data of Jůza (1963) display very different compressibilities from the others and are higher by 3 parts in 10^4 at 1000 bars and 0°C . The data of Kennedy et al. (1958) are lower by the same amount at 1000 bars and 0°C . The results of this study are every-

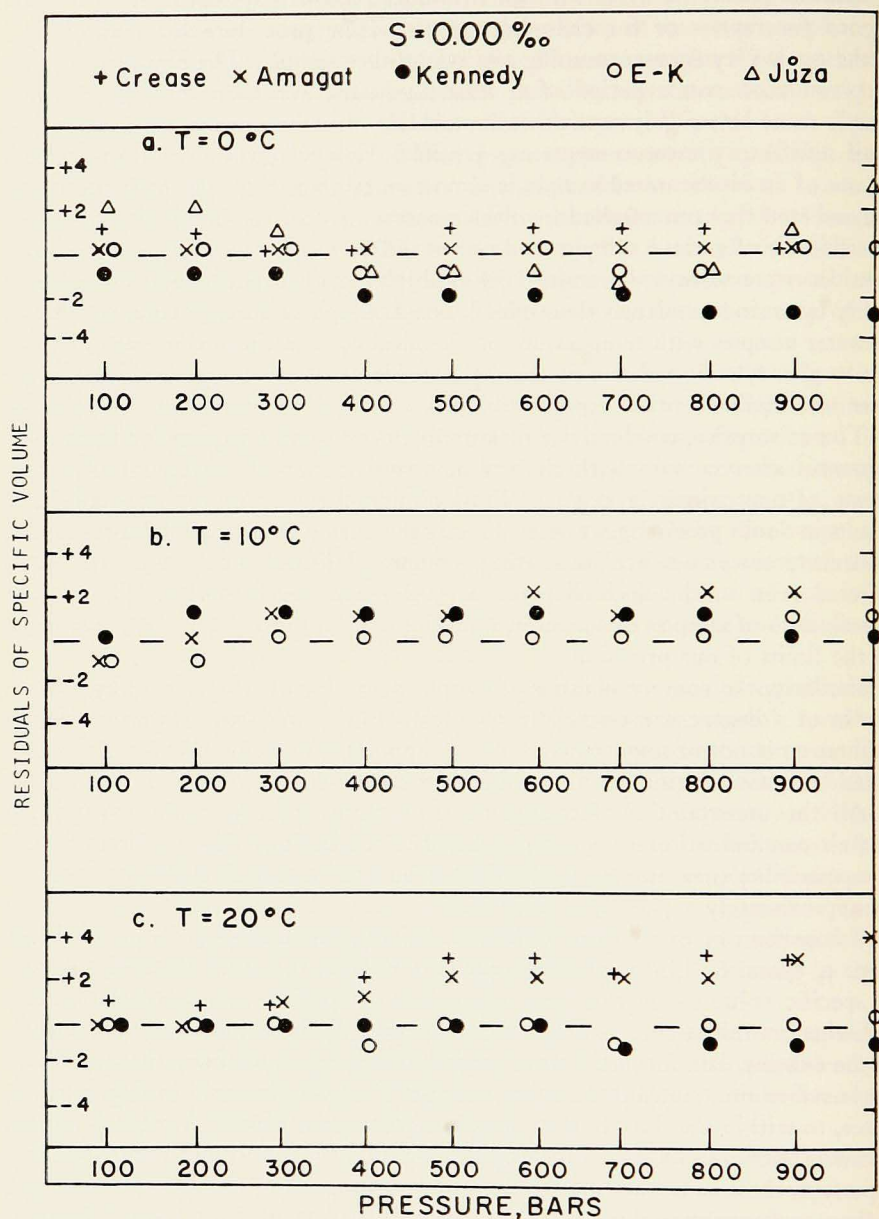


Figure 4. Comparison of results. Residuals are the values of specific volume of the other investigators minus those of our work times 104.

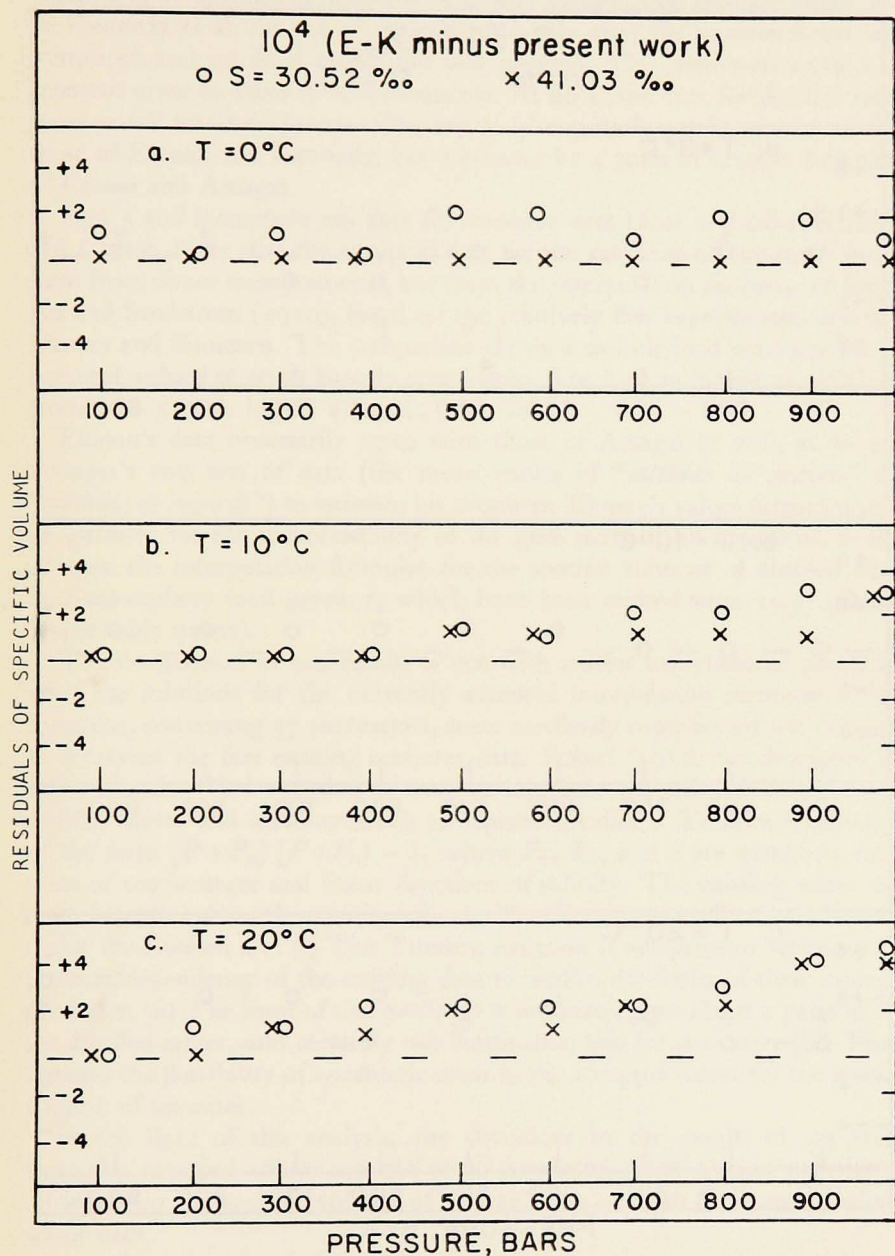


Figure 5. Comparison of results. Residuals are the values of specific volume of the other investigators minus those of our work times 10⁴.

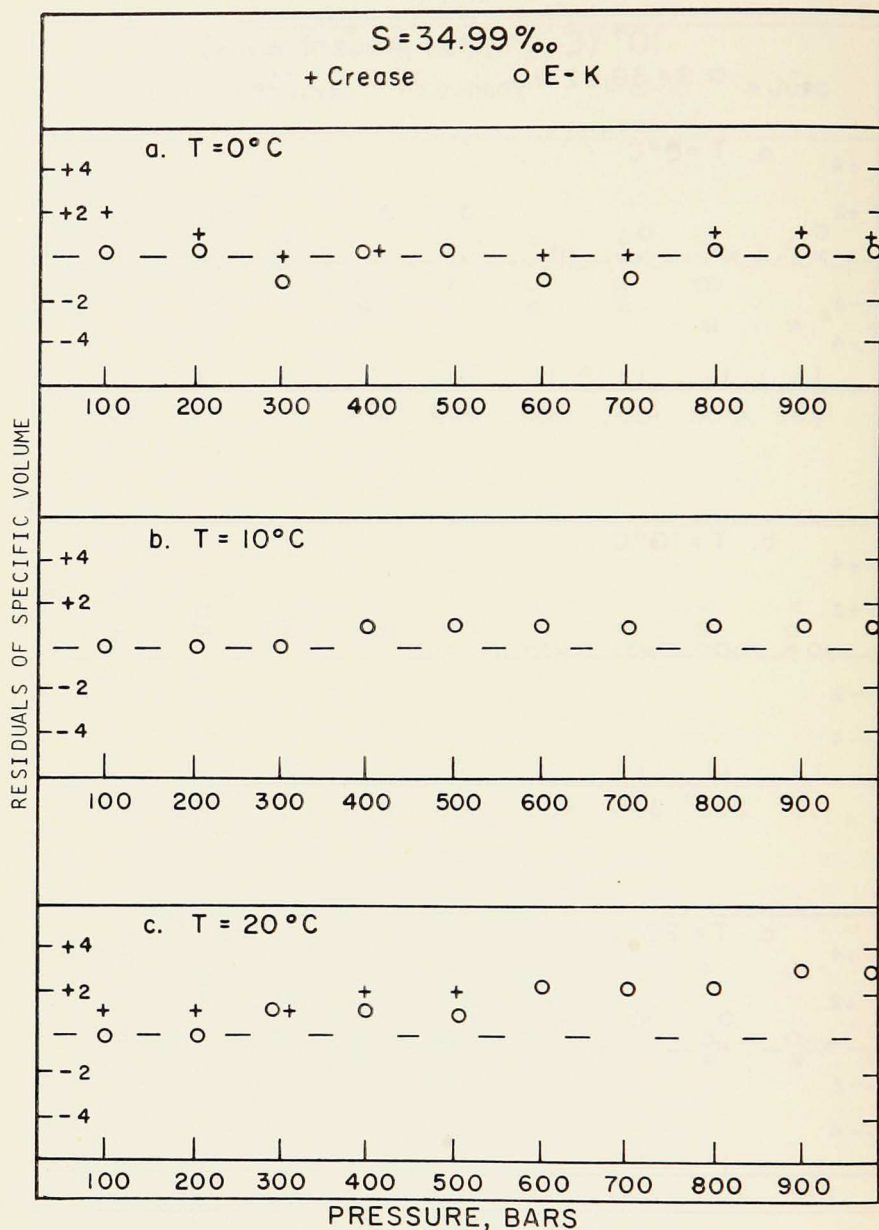


Figure 6. Comparison of results. Residuals are the values of specific volume of the other investigators minus those of our work times 104.

where else in agreement with the Kennedy values. The pressure gauge used by Kennedy et al. for the 0° data failed shortly after the measurements were completed and no final calibration was possible. This may well explain the apparent error in those 0° measurements. At 20°C the data for distilled water seem to fall into two groups. The values of our study are in agreement with those of Ekman and Kennedy, but are lower by 4 parts in 10,000 than those of Crease and Amagat.

Figs. 5 and 6 compare our data for seawater with those of Ekman-Knudsen and Crease. Note that the accepted data for the salinities of our study do not stem from direct measurements, but from the interpolation formulae of Bjerkenes and Sandstrom (1910), based on the relatively few experimental points of Ekman and Knudsen. The comparison shows a well-defined tendency for the accepted values at 1000 bars to range from 1 to 3 parts higher at 10°C and from 3 to 5 parts higher at 20°C.

Ekman's data necessarily agree with those of Amagat at 0°C, as he used Amagat's two sets of data (the mean values of "*méthode de contacts*" and "*méthode de regards*") to estimate his pressures. Ekman's values depended upon an estimate for the compressibility of the glass parts of his apparatus, as well as upon the interpolation formulae for the specific volumes of distilled water at 1-atmosphere total pressure, which have been revised since 1937 (Dorsey 1940: table 94-III).

The comparison of our results is not with similar experimental points but with the solutions for the currently accepted interpolation formulae. These formulae, containing 37 parameters, seem needlessly complicated and elaborate to represent the few existing seawater data. Eckart (1958) has developed the following simplified equation of state to represent the existing data for both distilled water and seawater. Such an equation, called a Tumlirz equation, is of the form $[P + P_0] [V + V_0] = \lambda$, where P_0 , V_0 , and λ are quadratic functions of temperature and linear functions of salinity. The existing water data seem to scatter about the solutions for the Tumlirz equation. Professor Eckart's major conclusions are: (i) The Tumlirz equation is adequate to represent the pressure dependency of the existing data to within the limits of their inherent precision; (ii) The limit of this precision is not better than about 2 parts in 104 for distilled water, and certainly not better than this for seawater; (iii) There remains the possibility of systematic error in the accepted values for the specific volume of seawater.

In the light of this analysis, the deviations in the results of our study from the accepted values are not at all surprising. Only further experimental work on the specific volume of seawater can establish the accuracy of any set of data.

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