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# A Vibrating Tube Flow Densitometer for Measurements with Corrosive Solutions at Temperatures up to 723 K and Pressures up to 40 MPa

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A new version of a vibrating tube flow densitometer has been designed permitting measurements of density differences between two fluids in the temperature range from 298 to 723 K and at pressures up to 40 MPa. The instrument is equipped with a Pt/Rh20 vibrating tube (1.6-mm o.d.) and a Pt/Rh10 transporting tube (1.2-mm o.d.) permitting measurements with highly corrosive liquids. The period of oscillation of the tube is about 7.5 ms, with a typical stability better than $10^{-4}\%$ over about a 1-h period over the entire temperature interval. The calibration constant $K$ at room temperature is about $530\ \mathrm{kg \cdot m^{-3} \cdot ms^{-2}}$, with a temperature coefficient of approximately $-0.13\ \mathrm{kg \cdot m^{-3} \cdot ms^{-2} \cdot K^{-1}}$, and is practically pressure independent. It can be determined by calibration with a reproducibility generally better than 0.1%. The instrument was tested with NaCl(aq) solutions in the temperature range from 373 to 690 K for density differences between sample and reference liquid ranging from 200 to $2\ \mathrm{kg \cdot m^{-3}}$; the corresponding errors are believed to be below 0.3 and 5%, respectively. A highly automated temperature control maintains the temperature of the tube stable to within ±0.02 K.

**KEY WORDS:** aqueous solutions; critical point flow densitometer; high temperature; high pressure; vibrating tube.

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## 1. INTRODUCTION

Vibrating tube flow densitometry has proven to be an efficient tool for studying the volumetric properties of solutions over a wide range of temperatures and pressures. In this approach the density of a sample is obtained by measurements on a stream of fluid introduced continuously at a low flow rate in a thin metallic tube vibrating in a magnetic field. The difference between the density of a sample $\rho$ and that of a reference fluid (usually water) $\rho_0$ is obtained by comparing vibration periods $\tau$ of the tube filled alternatively with a sample and a reference substance:

$$\Delta\rho = \rho - \rho_0 = K(\tau^2 - \tau_0^2) \tag{1}$$

where $K$ is a calibration constant obtained from an experiment with two fluids of well-known densities. The main advantages of this approach are fast temperature equilibration, no vapor space correction, and a low residence time, limiting tube corrosion and/or risk of sample decomposition. The method is particularly suitable for volumetric investigation of dilute solutions where most other techniques give less reliable results.

Commercial models of vibrating tube densitometers (Anton Paar) are based on the original design of Kratky et al. [1] and have a permanent magnet attached to a vibrating tube, which is located between two wire-coil electromagnets. Several nonflow set ups operating at temperatures not far removed from ambient and pressures up to 40 MPa were described recently in the literature [2–4]. An important quantity of papers reporting densities measured on vibrating tube densitometers appeared in the last five years or so, particularly in connection with research on new types of ecologically safe refrigerants. Commercial models do not, however, allow measurements above 423 K. A high-temperature flow densitometer using Anton Paar drive and sensing circuits was recently constructed at the Oak Ridge National Laboratory by Simonson et al. [5] and used for measurements with aqueous electrolytes [6] of ionic strength above 0.5 and temperatures up to 523 K.

Wood and collaborators [7, 8] at the University of Delaware have proposed an inverted design using a permanent horseshoe magnet and a vibrating tube carrying the drive and pickup rods attached in its upper third by special ceramics. The drive and pickup circuits are coupled by an electronic system using the principle of a phase locked loop which sustains vibration at the resonance frequency [9]. This instrument has yielded a considerable amount of data for aqueous electrolyte solutions up to 600 K (e.g., see Refs. 8, 10, and 11). The same principle was used in the densitometer by Corti et al. [12], which yielded data up to 523 K and concentrations above 0.5 m. A new type of vibrating tube, which oscillates in an overtone rather than in its fundamental mode, was described by Chang and Moldover [13]; this design avoids the use of electromagnets or appendages on the oscillating tube because the alternating drive current is passed directly through the tube vibrating in a field of a permanent magnet. A similar approach was adopted at the Institute of Chemical Technology in Prague [14], where a photoelectric signal is used as a pickup from a tube directly driven by a current.

Most of the densities in the literature obtained by vibrating tube densitometry are at temperatures below 523 K from instruments that usually do not allow investigation of highly dilute solutions and that often have limitations regarding the chemical character of the investigated substances. Our objective was to build an instrument that permits rapid measurements with corrosive aqueous solutions over a wide range of temperatures and pressures up to the so-called critical region of water (extending, say, 75 K and 15 MPa above the critical point of water and roughly symmetrical by the critical isochore $\rho_c = 322\ \mathrm{kg \cdot m^{-3}}$). The instrument should be sensitive enough to produce data for dilute solutions [near ionic strength of 0.01 molal for electrolytes(aq)]; the dependence of volume on concentration becomes very strong at high temperatures, and data in the low-concentration region ($m < 0.2\ \mathrm{kg \cdot m^3}$) are crucial for extrapolating to infinite dilution. The new densitometer at the University Blaise Pascal, described in this publication, is a modification and improvement of a design described previously by Majer et al. [15]. This design yielded practically all of the literature data for dilute aqueous solutions at temperatures above 573 K published to date [16–21]. In the new instrument the design of the vibrating tube unit has been modified and the densitometer features a better preheating system and an automated temperature control and data retrieval.

## 2. DENSITOMETER

### 2.1. Mechanical Parts

The vibrating tube assembly and the densitometer cross section are depicted in Figs. 1 and 2, respectively. The "U"-shaped vibrating tube (o.d., 1.6 mm; i.d., 1.1 mm; height, 72 mm; "U" radius, 12 mm) of platinum–rhodium alloy (20% of Rh) is located between the two permanent horseshoe magnets (active area, 30 × 11 mm) that are fixed in the aluminium densitometer block. The magnetic field is concentrated by an iron pole piece (active area, 18 × 16 mm) to the drive and pickup bars (tungsten wire; o.d., 0.8 mm; length, 30 mm), which are attached to the upper part of the tube by means of a special high-temperature-resistant insulating

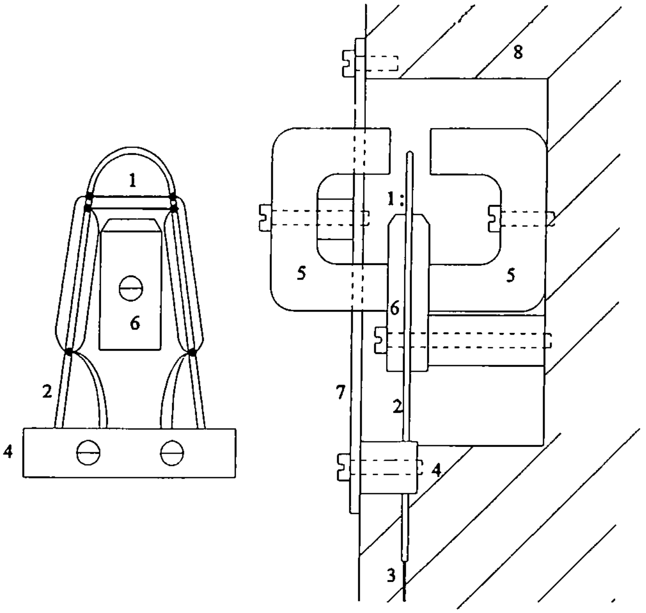

Fig. 1. Vibrating tube assembly: (1) Pickup and drive bars; (2) vibrating tube; o.d., 1.6 mm; (3) transporting tube; o.d., 1.1 mm; (4) brass block; (5) horseshoe magnet; (6) extension pole piece; (7) cover; (8) densitometer block.

ceramic (the distance between the pole piece and the lower bar is 2 to 3 mm). Thin electric leads (0.1 mm platinum wire) are silver-soldered to the ends of the bars and descend along the tube. After passing through the brass block to which the vibrating part is silver soldered, the diameter of the tubing is reduced to 1.2-mm o.d. (i.d. about 0.7 mm). Two branches of the thinner Pt/Rh tubing transport the fluid in and out of the vibrating tube.

The liquid coming into the densitometer is first thermostated in a countercurrent heat exchanger, formed by twisting the branches of the transport tubing together. Most of the heat exchanger is in close contact with a lid (diameter, 278 mm; thickness, 12 mm) heated by two strip heaters connected in parallel (each 200 W at 120 V). Before entering the block, the tubing supplying fluid to the vibrating tube passes through a preheater made from aluminium (length, 70 mm; cross section, 40 x 16 mm) equipped with a cartridge heater (o.d., 6 mm; length, 60 mm; 1 W at 20 V) and a platinum resistance thermometer. An iron/constantan thermocouple is used to detect any possible temperature difference between the inlet tube just above the top of the block and its lower part near the vibrating tube. This

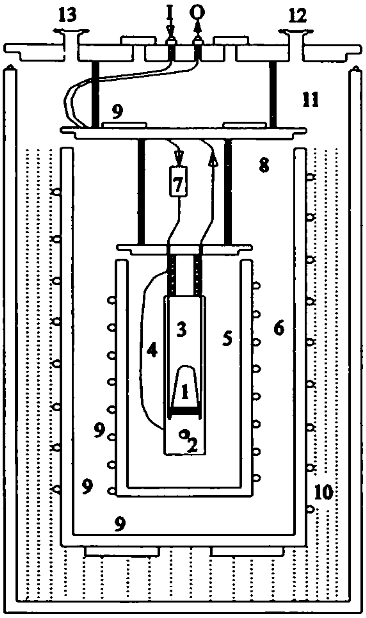

Fig. 2. Densitometer, cross-section view: (1) Vibrating tube; (2) thermometer; (3) densitometer block; (4) thermocouple; (5) inner can; (7) preheater; (8) lid with heaters; (9) heaters; (10) glass-wool insulations; (11) vacuum can; (12) connection to vacuum pump; (13) connection to vacuum gauge.

measurement allows adjustment of the preheater before starting experiments in order to have the voltage of the thermocouple close to zero.

The vibrating tube unit is housed in a cavity of a massive aluminium block (height, 325 mm; width 50 mm; depth, 75 mm) which is suspended on four threaded rods. The tubes that transport the fluid to and from the vibrating tube are located inside grooves on two opposite surfaces of the block. To provide a good thermal contact, the space between transporting tubes and the groove is packed with aluminium foil and tightly pressed against the block by bolted metallic plates. The block has a cartridge heater that can be used to quickly change the block temperature; however, during experiments the block is always used in a floating regime of temperature.

The block temperature is measured by a customized 500-$\Omega$ Burns Engineering secondary standard thermometer (6.0-mm o.d., 144-mm length) which has an accuracy of ±0.02 and ±0.033 K up to a temperature of 473 and 693 K, respectively. The thermometer is located inside the lower third of the block parallel to the "U"-vibrating tube. Due to possible temperature gradients, we expect that the temperature of the liquid entering the vibrating tube is known with an accuracy to ±0.05 K near room temperature and to ±0.2 K near the upper temperature limit. However, the uncertainty could become substantially higher for measurements near the critical point of water (approximately less than +20 K), where running an experiment in a flowing mode becomes difficult due to steep peaks in the compressibility and heat capacity of water observed near the critical isochore. These difficulties are attenuated as the distance from the critical point increases.

The densitometer block is surrounded by inner and outer cans (diameters, 165 and 278 mm; heights 420 and 800 mm, respectively) equipped with thermocoax heaters located in the grooves on the outer cylindrical surface (36 and 14 $\Omega$ and 250 and 750 W, respectively, at 110 V) and Pt thermometers. Their whole assembly is placed inside a vacuum envelope (o.d., 400 mm; height, 930 mm) containing glass–wool insulation that is maintained at a pressure of less than 25 Pa. The lid of the vacuum envelope is connected to the heated lid, the inner can lid, and the block by threaded rods in one mechanical unit which can be easily taken out of the densitometric can. The inner can is bolted to its lid and can be removed for accessing the block after the whole unit is taken out.

## 2.2. Vibrating Tube Electronics

An alternating current is supplied to the drive circuit from a voltage-controlled oscillator (VCO) across a power stage. The tube starts vibrating and an electric signal induced in the pickup bar is introduced to the phase comparator and compared with the drive signal. Either the VCO is controlled by the filtered output voltage from the phase comparator or the voltage is adjusted by means of a potentiometer (manual control when starting vibration). This is the principle of the phase locked loop operation [9], which sustains vibration at a resonance frequency (about 133 Hz, corresponding to a period close to 7.5 ms).

The pickup signal, which is closely linked with the vibration amplitude, tends to decrease with increasing temperature provided that the driving current remains constant. This is due to a combination of several factors playing a role at high temperatures including changes in mechanical properties of the tube, weakening of the field of the permanent magnets, and an increase in the electrical resistance in the drive circuit. With decreasing amplitude, the oscillation is less affected by mechanical damping effects, however, the pickup signal is prone to perturbations due to electromagnetic noise. This is the case particularly at high temperatures when the heaters on the outer can and on the lid are working with a high electric input. It is therefore necessary to find at each temperature an optimum vibration amplitude which is rather low but at which the frequency is reasonably stable and is not prone to perturbations from outside. For that reason the drive current is changed from about 20 mA near room temperature to 35 mA near the upper temperature limit. The estimated Joule effect in the two 0.1-mm Pt leads of the drive bar is generally below 1 mW close to ambient temperature and below 10 mW near the upper temperature limit. It is realistic to assume that a major part of this heat is lost to the brass block and the air, and only a minor part causes heating of the tube. We therefore expect that there is no danger of overheating causing a temperature increase in the tube.

The period of the vibration is calculated from a measured time interval corresponding to a selected number of oscillations (usually 2000, resulting in one reading about every 15 s). This time interval is measured and stored by means of a programmable counter 8254 placed on an "I/O" Selectronics card in a PC386. The integrated circuit 8254 contains three independent programmable counters. The first one counts pulses corresponding to oscillations of the vibrating tube and determines the time period during which the other two counters will be opened for counting the pulses of the clock. The latter two counters are connected in series to avoid an overflow due to insufficient capacity. Finally, the contents of the two counters (the time corresponding to a selected number of vibrating tube periods) is read by the computer and is further processed to obtain an averaged period of the vibrating tube. The software automatically checks the value of the period and is able to correct or simply exclude occasional spurious counts before storage in an output file on a disk.

### 2.3. Temperature Regulation

The main task of the temperature control system is to maintain the constancy of the temperature of the densitometer block and to control the temperature of the incoming liquid. The instrument is equipped with four regulation circuits; temperature controls of the inner and outer cans and of the preheater are independent, but the temperature of the lid is adjusted by using as reference that of the inner can. During experiments the temperature of the densitometer block remains floating to avoid temperature gradients. The outer can is at a temperature about 0.3 K lower than that of the inner can. The lid temperature is adjusted close to or above the inner can temperature to contribute significantly to the temperature control of the incoming fluid. The temperature of the fluid is brought to that of the block (±0.1 K) after passage through the low-wattage preheater; its temperature is adjusted manually to keep the signal from the thermocouple close to zero. The stability of the block temperature is generally better than ±0.01 K during one experiment, and the long-term drifts (say, over a 24-h period) are below ±0.05 K.

The structure of the temperature regulation is identical for all four control units and is depicted in Fig. 3. A Hartman–Braun platinum resistance thermometer (1000 $\Omega$, 3 mm o.d., 14-mm length for the inner and outer can regulations and 100 $\Omega$, 3-mm o.d.; 12-mm length for the lid and preheater regulations) and a resistor decade are connected in two branches of a Wheatstone bridge, which serves as a detector of a regulation deviation. In the case of the lid regulation, the decade is replaced by a second thermometer on the inner can; the temperature difference between the two units can be changed continuously from about −5 to +5 K with the aid of a helipot connected in parallel with a 20-$\Omega$ resistor located between the two fixed resistors of a Wheatstone bridge. All four bridges are supplied by direct current from a 5-V stabilized source. The output signals from the bridges are amplified (gains: 20,000 inner can, 10,000 outer can, 1000 lid, 8000 preheater) and fed across a multiplexer to a 12-bit A/D convertor (analog range from −4 to 4 V) on an "I/O" Selectronics card in a personal computer. After conversion, the digital information is processed by the regulation software, which adjusts the optimum regulation function. Owing to the isothermal regime of the instrument and strict requirements

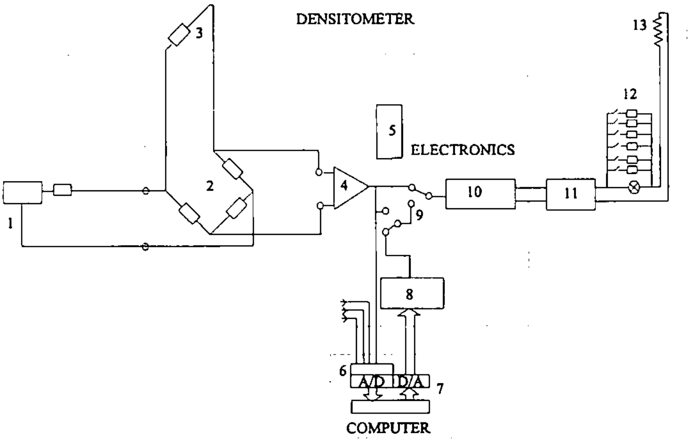

**Fig. 3.** Regulation structure. (1) Source; (2) Wheatstone bridge; (3) RTD (100 or 1000 $\Omega$); (4) amplifier; (5) security relays; (6) multiplexer; (7) control card; (8) multiplexer (sample and hold); (9) switch; (10) converter; (11) power pack; (12) set of parallel resistors; (13) heater.

on the high quality of the temperature stability, only proportional high-gain regulation was used. The signal from the computer is converted again to an analog signal (0–4 V) and amplified to a voltage of 0 to 10 V, which commands a 11906-223 power package made by Leeds and Northrup to supply electric energy to the heater.

Simplified regulation is also possible without using a computer and this regime is used mainly in case the software malfunctions or if there is a computer accident. In the normal software function an alternating current is produced during each regulation cycle, this current is redressed and filtered keeping the security relays connected. When the computer is disconnected or fails to function normally, this current is absent and the security relay automatically switches to regulation without a computer.

In the case of the outer can and lid regulation, the energy required depends largely on the operating temperature. A battery of variable parallel resistors connected in series with the heater allows us to adjust the amount of electric energy supplied according to the heating input needed.

## 3. EXPERIMENTAL

### 3.1. Operation of the Instrument

The overall schematic setup of the instrument is given in Fig. 4. The redistilled, deionized, degassed water can be introduced to the system by an isocratic or piston pump at a flow rate of about 0.4 to 0.5 $\mathrm{cm^3 min^{-1}}$. In our setup the pressure is kept constant to 0.01 MPa by means of a BPR21 series flow pressure regulator ($p < 40$ MPa) from Circle Seals Controls. The dome of the back pressure regulator is loaded with compressed nitrogen to achieve the experimental pressure. The regulator should be thermostated to avoid pressure drifts due to possible changes in the laboratory temperature. When measuring above the critical temperature of water under conditions near the critical isochore, it is advisable to use two regulators in series [15] (the second at a pressure about 1 MPa lower than the system pressure) to avoid pressure fluctuations due to the high compressibility of water. In this case the use of piston pumps which are pulse-free is preferable to working with reciprocating pumps, which are otherwise largely sufficient for delivering liquids outside the critical range.

The pressure in our test experiments (see below) was measured by a DPI 260 Druck electronic pressure gauge with an accuracy to 0.15%, which is sufficient for most measurements except in the vicinity of the critical point. Considering the equipment used for measuring and regulating pressure and temperature, it is not advisable to perform in the present configuration experiments close to the critical point of water. By this, we mean

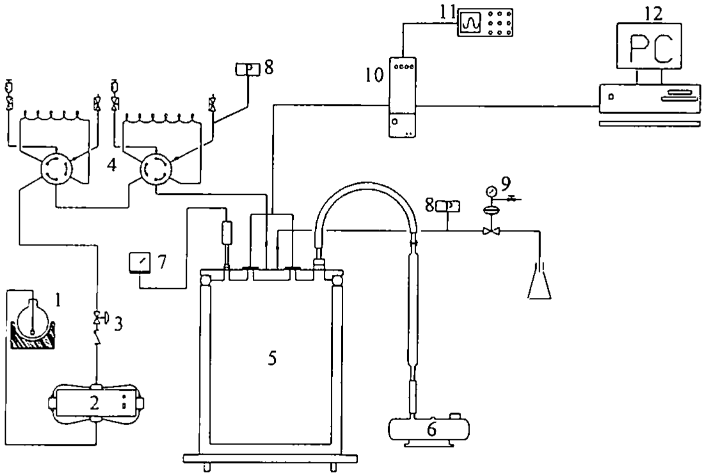

**Fig. 4.** Overall schematic setup. (1) Water reservoir; (2) HPLC pump; (3) check and security valves; (4) filling system with six port valves; (5) densitometer can; (6) vacuum pump; (7) vacuum gauge; (8) pressure indicators; (9) back pressure regulator; (10) electronic control unit; (11) oscilloscope; (12) computer.

the region between temperatures of 643 and 663 K at pressures from 22 to 27 Mpa. Fast variation of density and heat capacity with $T$ and $p$ in this region could seriously compromise the reliability of the results given the present accuracy of temperature and pressure determination and the stability of their maintenance. Upgrading of devices for measuring and controlling $T$ and $p$ would have only a limited effect, as flow methods are not generally applicable in the close vicinity of the critical point because of possible fluctuations in the system.

After degassing, the investigated solution is loaded into a sample loop which is first bypassed by water, entering directly into the densitometer. The averaged values of the vibration period (about 7.5 ms) are viewed on a computer screen, and when the period reading becomes stable to about 5 ns or exhibits only a minor and regular drift (the same sign differences of about 2 to 3 ns in two subsequent readings), it is recorded for a period of 10 to 20 min ("baseline" determination). After switching the six-port valve, water is introduced to the sample loop, forcing the solution from the loop into the densitometer. After about 10–15 min, the new signal becomes again stable and the "plateau" is registered for about 10 min. The experiment is completed by returning the valve to a bypass position and remeasuring the water base line. One data point can be obtained in 30 to 45 min (determinations for concentrated solutions take longer). When working under conditions where the compressibility of water is high, there is the possibility of pressure perturbations (and consequently also perturbation of the baseline and the plateau). To avoid this the sample loop should be prepressurized immediately before the sample injection.

The system is equipped with two sample loops to speed up the experiments. When working with samples immiscible with water, the first loop can be filled with a mutually soluble liquid which is introduced into the densitometer before and after the plateau to separate the water from the sample loaded to the second loop closer to the densitometer. In calibrations where water and a gas are used, it is convenient to fill the first loop with a volatile fluid which separates the gas from the water. In this way problems arising from incomplete drying of the tube, when the gas replaces the water, can be avoided. It was observed that small drops of water tend to stick on the internal tube wall at temperatures below 473 K and are only slowly eliminated by streaming gas.

It is convenient to use water and 3 molal NaCl(aq) or heavy water for calibrating the densitometer between room temperature and 373 or 473 K, respectively; operation is identical in this case as during a regular experiment. Highly accurate data (error in density below 0.01%) are available for both 3 M NaCl(aq) and $D_2O$ in the above temperature ranges and their density compared to that of water is about 10% higher which is enough to obtain the calibration constant with sufficient precision. A gas (usually nitrogen or helium) and water can be used starting at a temperature of 373 K. Since the difference between the density of water and that of gas is much larger, a better precision in the constant determination can be achieved. Helium is more advantageous to use at temperatures above 573 K, where the difference between water and nitrogen diminishes. However, it should be kept in mind that the calibration is performed under conditions which are rather different from those of a regular experiment with an aqueous solution and this can introduce a certain error. We have compared the calibration constants obtained in the two calibration regimes [NaCl(aq)/$H_2O$ and $N_2/H_2O$] at a temperature of 373 K. The difference was less than 0.2%, indicating a reasonably linear relationship between the density of a fluid and the second power of the vibration period. The calibration constant decreases with increasing temperature by about 15% over 400 K and shows only a small dependence on pressure. The reproducibility of the calibration constant determination is about 0.05%. The expected error in $K$ increases from about 0.1% near room temperature to about 0.5% near the upper temperature limit.

### 3.2. Tests

We have tested the densitometer by measuring densities of both dilute ($m \leqslant 0.1$ mol·kg$^{-1}$) and concentrated ($m \approx 1$ and 3 mol·kg$^{-1}$) solutions of NaCl(aq) between 373 and 573 K and at 10 and 30 MPa. To demonstrate the capability of the instrument to yield data at temperatures and pressures where the compressibility of water is large, we have performed additional experiments in the critical region of water along the isobar of 30.25 MPa. The maximum water compressibility ($\kappa_T \approx 0.14\ \mathrm{MPa}^{-1}$) at this pressure is near 678 K. Experiments could not be performed for the 3 molar solution at the highest temperature, because phase separation was observed (instability of the plateau, decrease of the vibration amplitude). For tests above 373 K the selected concentrations matched those of the earlier investigation of NaCl(aq) [17] in the critical region of water to allow direct comparison without interpolation.

Solutions were prepared by weight from a commercial sample of NaCl (Fluka; analytical grade reagent) dried for 24 h under vacuum at a temperature near 400 K. Redistilled, deionized, and degassed water was used in all the experiments and was introduced to the system by a Milton Roy Constametric I HPLC pump or, at the highest three temperatures, with a high-pressure piston pump (HPP 5001 of Laboratorni pristroje, Prague). Calibrations were performed with nitrogen and water at each combination of temperature and pressure. Operational mode of experiments and auxiliary equipments were as described above.

Table I lists the density differences $\Delta\rho$ and the apparent molar volumes $V_\phi$ of NaCl(aq) calculated from the equation

$$V_\phi = \frac{M_{\mathrm{NaCl}}}{\rho_{\mathrm{w}}} - \frac{1000\Delta\rho}{m\rho\rho_{\mathrm{w}}} \tag{2}$$

where the subscript w refers to water. The test results were compared with the literature values, which correspond to the recommendations published by Archer [22] at temperatures up to 573 K based on an evaluation of all thermodynamic data available for NaCl(aq). The literature data at higher temperatures are experimental values by Majer et al. [17] measured on a vibrating tube densitometer developed at the University of Delaware [15]. In the region of high dilution these data are the only results available in the literature above 573 K. The agreement with the literature data is in most cases better than 0.5 and 3% in $\Delta\rho$ at temperatures below and above 573 K, respectively. The uncertainly in $\Delta\rho$ is sensitively reflected, particularly in the apparent molar volumes at low concentrations (error is proportional to the reciprocal value of molality). The data of the test experiments reproduce well the minimum in the apparent molar volume of dilute NaCl(aq) as a function of temperature between 673 and 681 K. The presented comparisons prove that the new instrument is capable of yielding

**Table I.** Density Differences $\Delta\rho$ and Apparent Molar Volumes $V_\phi$ of NaCl(aq)

| $T$ (K) | $p$ (MPa) | $m$ (mol · kg$^{-1}$) | $\Delta\rho$(exp) (kg · m$^{-3}$) | $\Delta\rho$(lit) (kg · m$^{-3}$) | $V_\phi$(exp) (cm$^3$ · mol$^{-1}$) | $V_\phi$(lit) (cm$^3$ · mol$^{-1}$) |
|---|---|---|---|---|---|---|
| 374.05 | 10.2 | 0.1082 | 4.26 | 4.24 | 20.9 | 20.8[a] |
| 374.05 | 29.9 | 0.1082 | 4.19 | 4.17 | 18.8 | 19.1[a] |
| 374.05 | 29.9 | 2.9810 | 102.63 | 102.64 | 21.4 | 21.4[a] |
| 447.44 | 10.1 | 0.04996 | 2.24 | 2.22 | 9.6 | 10.0[a] |
| 447.43 | 10.1 | 0.9993 | 40.66 | 40.91 | 14.0 | 13.7[a] |
| 447.43 | 30.6 | 0.04996 | 2.17 | 2.15 | 11.6 | 12.3[a] |
| 447.43 | 30.5 | 0.9993 | 39.78 | 39.93 | 15.5 | 15.3[a] |
| 450.05 | 10.2 | 3.0114 | 112.71 | 112.70 | 16.6 | 16.5[a] |
| 450.07 | 30.8 | 3.0114 | 110.08 | 110.48 | 17.9 | 17.7[a] |
| 518.35 | 10.2 | 0.04996 | 2.86 | 2.86 | −14.6 | −14.6[a] |
| 518.36 | 10.2 | 0.9993 | 49.16 | 49.21 | −2.4 | −2.5[a] |
| 518.37 | 30.2 | 0.04996 | 2.65 | 2.65 | −6.4 | −6.2[a] |
| 518.38 | 30.2 | 0.9993 | 46.33 | 46.55 | 3.1 | 2.8[a] |
| 572.70 | 10.3 | 3.0114 | 161.04 | 162.80 | −18.4 | −19.3[a] |
| 572.66 | 30.2 | 3.0115 | 145.15 | 144.53 | −6.6 | −6.0[a] |
| 655.08 | 30.28 | 0.009831 | 2.27 | 2.25 | −720.5 | −709.6[b] |
| 655.08 | 30.29 | 2.9883 | 246.1 | 250.9 | −127.0 | −130.4[b] |
| 667.82 | 30.22 | 0.009831 | 4.05 | 4.08 | −2071 | −2090[b] |
| 667.82 | 30.22 | 2.9883 | 311.8 | 318.1 | −247.6 | −250.5[b] |
| 672.83 | 30.25 | 0.009831 | 5.22 | 4.95 | −3630 | −3420[b] |
| 672.83 | 30.25 | 2.9883 | 354.5 | 363.1 | −357.5 | −366.2[b] |
| 681.24 | 30.25 | 0.004981 | 1.33 | 1.34 | −3329 | −3270[b] |
| 681.24 | 30.25 | 0.02470 | 5.82 | 5.86 | −2869 | −2886[b] |

[a] From Ref. 22.
[b] From Ref. 17.

reliable data for aqueous solutions between near-ambient conditions and the critical region of water.

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