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# Pressure−Volume−Temperature Properties for 1-Octanol + Acetophenone, Poly(Propylene Glycol) + 1-Octanol + Acetophenone, and Poly(Ethylene Glycol) + Poly(Ethylene Glycol Methyl Ether) + Anisole

**Ming-Jer Lee, Tzu-Sheng Hsu, Yu-Chun Tuan, and Ho-mu Lin***

Department of Chemical Engineering, National Taiwan University of Science and Technology, 43 Keelung Road, Section 4, Taipei 106-07, Taiwan

Pressure−volume−temperature (*P*−*V*−*T*) properties were measured for 1-octanol + acetophenone, the polymer solutions of poly(propylene glycol) (PPG) with the mixed solvents of 1-octanol and acetophenone, and the polymer blends of poly(ethylene glycol) (PEG)/poly(ethylene glycol methyl ether) (PEGME) with anisole at temperatures from 298.15 K to 348.15 K and pressures up to 50 MPa. The Tait equation represents accurately the pressure effect on the liquid densities over the entire pressure range. The experimental-specific volumes of the constituent binary systems were correlated with the Flory−Orwoll−Vrij and the Schotte equations of state to determine the binary interaction parameters. By use of these determined binary parameters, the equations of state predicted the specific volumes to average absolute deviations of better than 0.13% for the ternary mixtures composed of PPG with the mixed solvents and the polymer blends of PEG/PEGME with anisole.

## Introduction

Reliable polymer equations of state (EOS) are useful in polymer manufacturing and processing as well as in many environmental and biological applications. In theory, the thermodynamic properties of a multicomponent system may be predicted from an EOS with known equation constants and binary interaction parameters, which are usually determined from the properties of the constituents and their binary pairs. The validity of the *P*−*V*−*T* relationship, describing by an EOS with mixing rules, can be tested directly with the experimental volumetric data of multicomponent systems.

While Zoller and Walsh[1] collected *P*−*V*−*T* properties for a wide variety of “pure” polymeric substances, the literature data are relatively scarce for polymer solutions and blends, especially for multicomponent systems. Muller and Rasmussen[2] and Sandell and Goring[3] measured the specific volumes for aqueous poly(ethylene glycol)s PEGs and for oligomeric propylene glycols. In the recent years, *P*−*V*−*T* data were reported for several associated polymer solutions and blends, such as poly(4-hydroxystyrene) + acetone (Compostizo et al.),[4] poly(4-hydroxystyrene) + tetrahydrofuran and + ethanol (Compostizo et al.),[5] poly(propylene glycol) PPG + *n*-hexane and + ethanol (Colin et al.),[6] and PEG + PPG (Colin et al.).[7] Panayiotou and Sanchez[8] correlated binary *P*−*V*−*T* data with the lattice−fluid EOS to examine the applicability of such models for representing the hydrogen-bonding effects on the volumetric properties. Our research group has also conducted a series of *P*−*V*−*T* measurements for various polymer solutions and blends containing PEG, PPG, and poly(ethylene glycol methyl ether) PEGME. Lee et al.[9] reported the volumetric properties for PEGME-350, PEG-200, PEG-600, and the blended mixtures of PEGME-350 with either PEG-200 or PEG-600, Chang et al.[10] for 10 fractionation cuts of PEG and PPG, Lee et al.[11] for PEG-200 + 1-octanol and PEG-600 + 1-octanol, Lee et al.[12] for PEGME-350 + anisole and PEG-200 + anisole, Lin et al.[13] for the binary systems of PPG-4000 with 1-octanol and acetophenone, and Lee et al.[14] for PPG-425 + anisole, PPG-425 + PEGME-350, PPG-425 + PEGME-350 + anisole, and PPG-200 + PPG-425 + anisole. In the present study, the *P*−*V*−*T* properties were measured for 1-octanol + acetophenone, the polymer solutions of PPG-4000 with the mixed solvents of 1-octanol and acetophenone, and the polymer blends of PEG-200 and PEGME-350 with anisole. The experimental results reveal the volumetric behavior for these associated polymer solutions in response to the effects of temperature, pressure, and composition. The ternary *P*−*V*−*T* data were also used to examine the validity of the prediction from the polymer EOS and Flory−Orwoll−Vrij (FOV) and Schotte equations with the binary interaction parameters determined from the constituent binaries.

## Experimental Section

Anisole (99+ mass %), 1-octanol (99+ mass %), acetophenone (99 mass %), and the fractionation cuts of PPG-4000, PEG-200, and PEGME-350 were supplied by Aldrich Chemical Co. (USA). The number-average molecular weights ($M_n$) and the polydispersities ($M_w/M_n$) were measured with a matrix-assisted laser desorption/ionization time-of-flight. The values are approximately 4960 and 1.008 for PPG-4000, 260 and 1.0742 for PEG-200, and 366 and 1.0188 for PEGME-350. Each component has been degassed by heat accompanying with agitation before use. The schematic diagram of the *P*−*V*−*T* apparatus has been illustrated by Lee et al.[12] Liquid mixture sample was prepared from the degassed compounds by mass to an uncertainty of ±0.0001 in mass fraction. The density was measured with a high-pressure densitometer (DMA 512 P, Anton Paar). Pressure

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* To whom correspondence may be addressed. Tel: +886-2-2737-6643. Fax: +886-2-2737-6644. E-mail: hml@ch.ntust.edu.tw.

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**Table 1. $P-V-T$ Properties of 1-Octanol (1) + Acetophenone (2)**

| | 298.15 K | | 318.15 K | | 348.15 K | | | 298.15 K | | 318.15 K | | 348.15 K | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $P$ | $\rho$ | $\kappa_T \times 10^4$ | $\rho$ | $\kappa_T \times 10^4$ | $\rho$ | $\kappa_T \times 10^4$ | P | $\rho$ | $\kappa_T \times 10^4$ | $\rho$ | $\kappa_T \times 10^4$ | $\rho$ | $\kappa_T \times 10^4$ |
| MPa | g·cm$^{-3}$ | MPa$^{-1}$ | g·cm$^{-3}$ | MPa$^{-1}$ | g·cm$^{-3}$ | MPa$^{-1}$ | MPa | g·cm$^{-3}$ | MPa$^{-1}$ | g·cm$^{-3}$ | MPa$^{-1}$ | g·cm$^{-3}$ | MPa$^{-1}$ |
| $w_1 = 0.1075$ ($x_1 = 0.1000$) | | | | | | | | | | | | | |
| 0.1 | 0.9960 | 5.861 | 0.9789 | 6.656 | 0.9525 | 7.859 | 30 | 1.0123 | 5.068 | 0.9970 | 5.531 | 0.9731 | 6.504 |
| 10 | 1.0017 | 5.571 | 0.9853 | 6.234 | 0.9597 | 7.348 | 35 | 1.0149 | 4.957 | 0.9995 | 5.380 | 0.9761 | 6.323 |
| 15 | 1.0045 | 5.436 | 0.9883 | 6.041 | 0.9632 | 7.116 | 40 | 1.0173 | 4.851 | 1.0022 | 5.238 | 0.9792 | 6.153 |
| 20 | 1.0072 | 5.308 | 0.9913 | 5.860 | 0.9666 | 6.899 | 45 | 1.0197 | 4.750 | 1.0048 | 5.104 | 0.9820 | 5.992 |
| 25 | 1.0098 | 5.185 | 0.9941 | 5.691 | 0.9699 | 6.695 | 50 | 1.0221 | 4.653 | 1.0073 | 4.977 | 0.9848 | 5.840 |
| $w_1 = 0.2132$ ($x_1 = 0.2000$) | | | | | | | | | | | | | |
| 0.1 | 0.9712 | 6.111 | 0.9545 | 6.983 | 0.9284 | 8.377 | 30 | 0.9877 | 5.230 | 0.9727 | 5.699 | 0.9492 | 6.612 |
| 10 | 0.9770 | 5.787 | 0.9609 | 6.495 | 0.9359 | 7.692 | 35 | 0.9903 | 5.108 | 0.9754 | 5.530 | 0.9523 | 6.390 |
| 15 | 0.9798 | 5.636 | 0.9640 | 6.275 | 0.9394 | 7.389 | 40 | 0.9928 | 4.992 | 0.9780 | 5.372 | 0.9553 | 6.183 |
| 20 | 0.9825 | 5.493 | 0.9670 | 6.069 | 0.9428 | 7.109 | 45 | 0.9951 | 4.881 | 0.9807 | 5.224 | 0.9582 | 5.989 |
| 25 | 0.9852 | 5.359 | 0.9699 | 5.878 | 0.9461 | 6.851 | 50 | 0.9975 | 4.776 | 0.9832 | 5.083 | 0.9610 | 5.808 |
| $w_1 = 0.3172$ ($x_1 = 0.3000$) | | | | | | | | | | | | | |
| 0.1 | 0.9479 | 6.386 | 0.9313 | 7.215 | 0.9057 | 8.509 | 30 | 0.9647 | 5.395 | 0.9497 | 5.913 | 0.9267 | 6.922 |
| 10 | 0.9538 | 6.018 | 0.9378 | 6.721 | 0.9131 | 7.904 | 35 | 0.9672 | 5.260 | 0.9524 | 5.741 | 0.9298 | 6.715 |
| 15 | 0.9566 | 5.848 | 0.9409 | 6.498 | 0.9167 | 7.632 | 40 | 0.9697 | 5.132 | 0.9551 | 5.580 | 0.9329 | 6.521 |
| 20 | 0.9594 | 5.688 | 0.9439 | 6.290 | 0.9201 | 7.379 | 45 | 0.9722 | 5.010 | 0.9577 | 5.428 | 0.9359 | 6.339 |
| 25 | 0.9620 | 5.538 | 0.9469 | 6.095 | 0.9235 | 7.143 | 50 | 0.9746 | 4.894 | 0.9603 | 5.285 | 0.9386 | 6.166 |
| $w_1 = 0.4195$ ($x_1 = 0.4000$) | | | | | | | | | | | | | |
| 0.1 | 0.9262 | 6.734 | 0.9100 | 7.516 | 0.8847 | 9.110 | 30 | 0.9432 | 5.522 | 0.9286 | 6.111 | 0.9062 | 7.070 |
| 10 | 0.9322 | 6.275 | 0.9166 | 6.981 | 0.8924 | 8.309 | 35 | 0.9458 | 5.362 | 0.9314 | 5.928 | 0.9093 | 6.818 |
| 15 | 0.9351 | 6.067 | 0.9197 | 6.740 | 0.8960 | 7.958 | 40 | 0.9483 | 5.212 | 0.9340 | 5.755 | 0.9123 | 6.584 |
| 20 | 0.9379 | 5.873 | 0.9228 | 6.515 | 0.8995 | 7.637 | 45 | 0.9507 | 5.070 | 0.9368 | 5.594 | 0.9153 | 6.367 |
| 25 | 0.9406 | 5.692 | 0.9257 | 6.306 | 0.9029 | 7.342 | 50 | 0.9532 | 4.937 | 0.9393 | 5.442 | 0.9182 | 6.164 |
| $w_1 = 0.5201$ ($x_1 = 0.5000$) | | | | | | | | | | | | | |
| 0.1 | 0.9059 | 6.944 | 0.8900 | 7.676 | 0.8651 | 9.356 | 30 | 0.9230 | 5.673 | 0.9087 | 6.304 | 0.8866 | 7.243 |
| 10 | 0.9119 | 6.461 | 0.8967 | 7.157 | 0.8728 | 8.525 | 35 | 0.9256 | 5.507 | 0.9115 | 6.123 | 0.8896 | 6.982 |
| 15 | 0.9149 | 6.244 | 0.8998 | 6.921 | 0.8765 | 8.162 | 40 | 0.9281 | 5.350 | 0.9142 | 5.952 | 0.8928 | 6.742 |
| 20 | 0.9176 | 6.040 | 0.9029 | 6.702 | 0.8800 | 7.829 | 45 | 0.9306 | 5.202 | 0.9169 | 5.792 | 0.8959 | 6.519 |
| 25 | 0.9203 | 5.851 | 0.9059 | 6.497 | 0.8834 | 7.525 | 50 | 0.9330 | 5.063 | 0.9195 | 5.640 | 0.8987 | 6.310 |
| $w_1 = 0.6192$ ($x_1 = 0.6000$) | | | | | | | | | | | | | |
| 0.1 | 0.8866 | 7.139 | 0.8711 | 8.194 | 0.8469 | 9.595 | 30 | 0.9038 | 5.824 | 0.8900 | 6.309 | 0.8685 | 7.470 |
| 10 | 0.8927 | 6.639 | 0.8778 | 7.451 | 0.8546 | 8.762 | 35 | 0.9064 | 5.651 | 0.8927 | 6.078 | 0.8717 | 7.206 |
| 15 | 0.8956 | 6.413 | 0.8811 | 7.126 | 0.8583 | 8.396 | 40 | 0.9090 | 5.491 | 0.8954 | 5.865 | 0.8748 | 6.962 |
| 20 | 0.8984 | 6.203 | 0.8841 | 6.830 | 0.8618 | 8.061 | 45 | 0.9114 | 5.338 | 0.8981 | 5.666 | 0.8778 | 6.735 |
| 25 | 0.9012 | 6.008 | 0.8871 | 6.559 | 0.8653 | 7.754 | 50 | 0.9138 | 5.194 | 0.9007 | 5.482 | 0.8807 | 6.523 |
| $w_1 = 0.7166$ ($x_1 = 0.7000$) | | | | | | | | | | | | | |
| 0.1 | 0.8686 | 7.343 | 0.8534 | 8.293 | 0.8296 | 10.029 | 30 | 0.8859 | 5.966 | 0.8724 | 6.541 | 0.8513 | 7.553 |
| 10 | 0.8747 | 6.818 | 0.8602 | 7.613 | 0.8374 | 9.038 | 35 | 0.8885 | 5.787 | 0.8752 | 6.321 | 0.8545 | 7.258 |
| 15 | 0.8776 | 6.582 | 0.8633 | 7.311 | 0.8411 | 8.612 | 40 | 0.8910 | 5.618 | 0.8779 | 6.115 | 0.8576 | 6.986 |
| 20 | 0.8804 | 6.362 | 0.8664 | 7.034 | 0.8446 | 8.225 | 45 | 0.8935 | 5.460 | 0.8805 | 5.923 | 0.8605 | 6.735 |
| 25 | 0.8832 | 6.158 | 0.8695 | 6.778 | 0.8480 | 7.874 | 50 | 0.8959 | 5.311 | 0.8831 | 5.744 | 0.8634 | 6.503 |
| $w_1 = 0.8126$ ($x_1 = 0.8000$) | | | | | | | | | | | | | |
| 0.1 | 0.8518 | 7.549 | 0.8370 | 8.509 | 0.8137 | 10.152 | 30 | 0.8692 | 6.094 | 0.8560 | 6.681 | 0.8354 | 7.734 |
| 10 | 0.8579 | 6.992 | 0.8439 | 7.797 | 0.8215 | 9.192 | 35 | 0.8718 | 5.906 | 0.8589 | 6.452 | 0.8386 | 7.442 |
| 15 | 0.8609 | 6.743 | 0.8470 | 7.482 | 0.8252 | 8.775 | 40 | 0.8742 | 5.729 | 0.8616 | 6.239 | 0.8417 | 7.172 |
| 20 | 0.8637 | 6.511 | 0.8502 | 7.194 | 0.8287 | 8.396 | 45 | 0.8768 | 5.564 | 0.8643 | 6.041 | 0.8446 | 6.922 |
| 25 | 0.8665 | 6.295 | 0.8532 | 6.927 | 0.8321 | 8.051 | 50 | 0.8792 | 5.409 | 0.8668 | 5.855 | 0.8475 | 6.690 |
| $w_1 = 0.9070$ ($x_1 = 0.9000$) | | | | | | | | | | | | | |
| 0.1 | 0.8360 | 7.695 | 0.8216 | 8.648 | 0.7984 | 10.647 | 30 | 0.8534 | 6.184 | 0.8406 | 6.790 | 0.8204 | 7.862 |
| 10 | 0.8421 | 7.115 | 0.8283 | 7.924 | 0.8064 | 9.519 | 35 | 0.8559 | 5.990 | 0.8434 | 6.558 | 0.8236 | 7.538 |
| 15 | 0.8451 | 6.855 | 0.8316 | 7.605 | 0.8102 | 9.040 | 40 | 0.8585 | 5.808 | 0.8461 | 6.342 | 0.8267 | 7.241 |
| 20 | 0.8480 | 6.616 | 0.8347 | 7.311 | 0.8138 | 8.608 | 45 | 0.8610 | 5.638 | 0.8488 | 6.140 | 0.8297 | 6.967 |
| 25 | 0.8507 | 6.392 | 0.8377 | 7.041 | 0.8172 | 8.217 | 50 | 0.8633 | 5.477 | 0.8513 | 5.951 | 0.8325 | 6.714 |

in the measuring cell was manipulated by a hand pump and monitored by a pressure transducer (Model-PDCR 911, 0-70 MPa, Druck) with a digital indicator (model-DPI 261, Druck). Pressure measurements were accurate to ±0.1 % at pressures higher than atmospheric. A thermostatic bath with circulating water regulated the temperature of the measuring cell to within ±0.03 K. A precision digital thermometer (model-1560, Hart Scientific) incorporated with a thermistor probe measured the bath temperature to an accuracy of ±0.02 K, and its uncertainty is about ±0.01 K. The oscillation period ($t_i$) of sample $i$ in the vibrating U tube was read with a densimeter of DMA 48 (Anton Paar) which was converted into density ($\rho_i$) via

$$\rho_i = A(t_i^2 - B) \tag{1}$$

where $A$ and $B$ are apparatus parameters determined by using the literature density data of two calibration fluids: pure water[15] and dry nitrogen.[16] The calibration was made at each temperature of interest over a pressure range (0.1−50) MPa. The calibration reproduced water densities with an average absolute deviation of 0.01% over the entire calibrated conditions. The viscosity differences between the samples and the calibration fluids might affect the accuracy

**Table 2. Correlated Results of the Tait Equation for 1-Octanol (1) + Acetophenone (2)**

| $T$/K | $w_1$[a] | $x_1$[b] | C | $D$/MPa | $\pi_1 \times 10^5$ [c] |
|---|---|---|---|---|---|
| 298.15 | 0.1075 | 0.1000 | 0.09993 | 170.40 | 6.05 |
| | 0.2132 | 0.2000 | 0.09704 | 158.70 | 6.78 |
| | 0.3172 | 0.3000 | 0.09330 | 146.00 | 3.38 |
| | 0.4195 | 0.4000 | 0.08323 | 123.50 | 2.25 |
| | 0.5201 | 0.5000 | 0.08402 | 120.90 | 2.01 |
| | 0.6192 | 0.6000 | 0.08552 | 119.70 | 3.86 |
| | 0.7166 | 0.7000 | 0.08599 | 117.00 | 2.33 |
| | 0.8126 | 0.8000 | 0.08546 | 113.10 | 3.10 |
| | 0.9070 | 0.9000 | 0.08518 | 110.60 | 2.66 |
| 318.15 | 0.1075 | 0.1000 | 0.08826 | 132.50 | 5.09 |
| | 0.2132 | 0.2000 | 0.08394 | 120.10 | 1.56 |
| | 0.3172 | 0.3000 | 0.08831 | 122.30 | 1.75 |
| | 0.4195 | 0.4000 | 0.08809 | 117.10 | 2.40 |
| | 0.5201 | 0.5000 | 0.09434 | 122.80 | 6.70 |
| | 0.6192 | 0.6000 | 0.07495 | 91.37 | 5.78 |
| | 0.7166 | 0.7000 | 0.08376 | 100.90 | 1.98 |
| | 0.8126 | 0.8000 | 0.08407 | 98.70 | 1.39 |
| | 0.9070 | 0.9000 | 0.08533 | 98.57 | 2.35 |
| 348.15 | 0.1075 | 0.1000 | 0.10020 | 127.40 | 8.86 |
| | 0.2132 | 0.2000 | 0.08478 | 101.10 | 2.47 |
| | 0.3172 | 0.3000 | 0.09870 | 115.90 | 6.25 |
| | 0.4195 | 0.4000 | 0.08517 | 93.39 | 2.15 |
| | 0.5201 | 0.5000 | 0.08641 | 92.26 | 4.96 |
| | 0.6192 | 0.6000 | 0.09037 | 94.08 | 2.59 |
| | 0.7166 | 0.7000 | 0.08267 | 82.33 | 4.76 |
| | 0.8126 | 0.8000 | 0.08726 | 85.85 | 2.80 |
| | 0.9070 | 0.9000 | 0.08131 | 76.27 | 3.28 |

[a] $w_1$ is the mass fraction of component 1. [b] $x_1$ is the mol fraction of component 1, calculated with the molecular weights of 130.231 and 120.151 for 1-octanol and acetophenone, respectively. [c] $\pi_1$ is defined as in eq 3.

of density measurement by an oscillating densitometer (Ashcroft et al.),[17] but the effect is generally minor. The accuracy of the density measurements, without the correction of viscosity effect, is estimated to within ±0.1%, and the uncertainty of the reported densities is about ±0.0001 g·cm$^{-3}$.

## Results and Discussion

***Binary System.*** The experimental densities of the constituent compounds (PEG-200, PEGME-350, PPG-4000, anisole, 1-octanol, and acetophenone) have been reported and compared with literature values in Lee et al.[12] and Lin et al.[13] The agreement is within the uncertainty of the measurements. Table 1 lists the measured density data for the binary mixed solvents of 1-octanol + acetophenone at temperatures from 298.15 K to 348.15 K and pressures up to 50 MPa. These data will be used to determine the binary interaction parameter for the pair of mixed solvents.

The pressure effect on the isothermal densities of a given composition can be represented accurately by the Tait equation

$$\frac{\rho - \rho_o}{\rho} = C \ln\left(\frac{D + P}{D + 0.1}\right) \tag{2}$$

where $\rho_o$ is the density at 0.1 MPa. The optimized values of $C$ and $D$ were obtained by fitting the Tait equation to the density data with the following objective function ($\pi_1$)

$$\pi_1 = \left[\sum_{k=1}^{n} |\rho_{k,\mathrm{calc}} - \rho_{k,\mathrm{exp}}|/\rho_{k,\mathrm{exp}}\right]/n \tag{3}$$

where $n$ is the number of data points. $\rho_{k,\mathrm{calc}}$ and $\rho_{k,\mathrm{exp}}$ refer to the calculated and the experimental densities for the $k$th point, respectively. Table 2 reports the calculated results for 1-octanol + acetophenone. With the values of $\rho_o$, $C$, and $D$, the Tait equation reproduces the densities at pressures higher than 0.1 MPa to within the experimental uncertainty. The isothermal compressibility $\kappa_T$ at given temperature, pressure, and composition can be calculated from its definition with the aid of the Tait equation

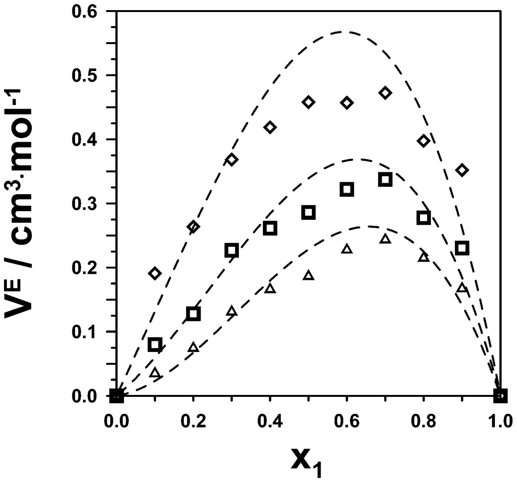

**Figure 1.** Variations of excess volume with composition for 1-octanol (1) + acetophenone (2) under 0.1 MPa at: ◇, 298.15 K; □, 318.15 K; △, 348.15 K.

**Table 3. Correlated Results of Excess Volumes with the Redlich–Kister Equation for 1-Octanol (1) + Acetophenone (2)**

| $T$/K | $P$/MPa | $E_1$/cm$^3$·mol$^{-1}$ | $E_2$/cm$^3$·mol$^{-1}$ | $\pi_2$[a]/cm$^3$·mol$^{-1}$ |
|---|---|---|---|---|
| 298.15 | 0.1 | 0.9131 | 0.8205 | 0.019 |
| | 10 | 0.9415 | 0.8065 | 0.022 |
| | 15 | 0.9799 | 0.8676 | 0.027 |
| | 20 | 0.9937 | 0.8042 | 0.024 |
| | 25 | 0.9572 | 0.7437 | 0.022 |
| | 30 | 0.9630 | 0.6877 | 0.023 |
| | 35 | 0.9813 | 0.7836 | 0.027 |
| | 40 | 1.0015 | 0.7444 | 0.026 |
| | 45 | 0.9919 | 0.7313 | 0.026 |
| | 50 | 0.9531 | 0.7262 | 0.028 |
| 318.15 | 0.1 | 1.3549 | 0.8793 | 0.026 |
| | 10 | 1.6163 | 1.2036 | 0.046 |
| | 15 | 1.6198 | 1.2163 | 0.045 |
| | 20 | 1.6011 | 1.1308 | 0.043 |
| | 25 | 1.6349 | 1.1242 | 0.046 |
| | 30 | 1.6286 | 1.2165 | 0.046 |
| | 35 | 1.6081 | 1.0391 | 0.044 |
| | 40 | 1.6390 | 1.0508 | 0.046 |
| | 45 | 1.6308 | 1.0873 | 0.046 |
| | 50 | 1.5985 | 1.1393 | 0.050 |
| 348.15 | 0.1 | 2.1830 | 0.9020 | 0.058 |
| | 10 | 2.0885 | 0.6387 | 0.049 |
| | 15 | 2.0144 | 0.5810 | 0.044 |
| | 20 | 2.0119 | 0.5133 | 0.041 |
| | 25 | 2.0260 | 0.5250 | 0.045 |
| | 30 | 2.0191 | 0.5784 | 0.045 |
| | 35 | 2.0275 | 0.5541 | 0.045 |
| | 40 | 1.9756 | 0.5551 | 0.042 |
| | 45 | 1.9671 | 0.5345 | 0.047 |
| | 50 | 1.9458 | 0.5155 | 0.046 |

[a] $\pi_2$ is defined as in eq 8.

$$\kappa_T = \frac{-1}{V}\left(\frac{\partial V}{\partial P}\right)_{T,x} = \frac{V_o}{V}\left(\frac{C}{D + P}\right) \tag{4}$$

where $V_o$ is the specific volume at 0.1 MPa. Table 1 lists the calculated isothermal compressibility at each experimental condition. It was found that the isothermal compressibility increases as the temperature is increased but decreases as the pressure is increased.

**Table 4. *P*−*V*−*T* Properties of PPG-4000 (1) + Acetophenone (2) + 1-Octanol (3)**

| | $\rho$/g·cm$^{-3}$ | | | $\rho$/g·cm$^{-3}$ | | |
|---|---|---|---|---|---|---|
| *P*/MPa | 298.15 K | 318.15 K | 348.15 K | 298.15 K | 318.15 K | 348.15 K |
| | $w_1/w_2/w_3$ = 0.8877/0.0400/0.0723<br>($x_1/x_2/x_3$ = 0.168/0.312/0.520) | | | $w_1/w_2/w_3$ = 0.8897/0.0668/0.0435<br>($x_1/x_2/x_3$ = 0.168/0.520/0.312) | | |
| 0.1 | 0.9844 | 0.9693 | 0.9460 | 0.9909 | 0.9753 | 0.9521 |
| 10 | 0.9906 | 0.9761 | 0.9537 | 0.9969 | 0.9820 | 0.9598 |
| 15 | 0.9936 | 0.9792 | 0.9573 | 0.9999 | 0.9853 | 0.9634 |
| 20 | 0.9964 | 0.9823 | 0.9609 | 1.0027 | 0.9883 | 0.9669 |
| 25 | 0.9992 | 0.9854 | 0.9643 | 1.0055 | 0.9914 | 0.9703 |
| 30 | 1.0020 | 0.9883 | 0.9675 | 1.0082 | 0.9943 | 0.9736 |
| 35 | 1.0047 | 0.9911 | 0.9707 | 1.0108 | 0.9972 | 0.9767 |
| 40 | 1.0072 | 0.9939 | 0.9738 | 1.0134 | 0.9999 | 0.9798 |
| 45 | 1.0098 | 0.9966 | 0.9768 | 1.0159 | 1.0027 | 0.9828 |
| 50 | 1.0122 | 0.9992 | 0.9797 | 1.0184 | 1.0053 | 0.9857 |
| | $w_1/w_2/w_3$ = 0.9540/0.0072/0.0388<br>($x_1/x_2/x_3$ = 0.350/0.108/0.542) | | | $w_1/w_2/w_3$ = 0.9552/0.0215/0.0233<br>($x_1/x_2/x_3$ = 0.350/0.325/0.325) | | |
| 0.1 | 0.9904 | 0.9752 | 0.9523 | 0.9939 | 0.9786 | 0.9557 |
| 10 | 0.9966 | 0.9820 | 0.9600 | 1.0001 | 0.9854 | 0.9634 |
| 15 | 0.9996 | 0.9852 | 0.9637 | 1.0031 | 0.9886 | 0.9671 |
| 20 | 1.0025 | 0.9884 | 0.9672 | 1.0060 | 0.9918 | 0.9706 |
| 25 | 1.0053 | 0.9914 | 0.9706 | 1.0087 | 0.9948 | 0.9739 |
| 30 | 1.0080 | 0.9944 | 0.9739 | 1.0115 | 0.9978 | 0.9773 |
| 35 | 1.0106 | 0.9972 | 0.9771 | 1.0141 | 1.0006 | 0.9804 |
| 40 | 1.0132 | 1.0000 | 0.9801 | 1.0167 | 1.0034 | 0.9835 |
| 45 | 1.0158 | 1.0027 | 0.9831 | 1.0193 | 1.0061 | 0.9865 |
| 50 | 1.0182 | 1.0054 | 0.9860 | 1.0217 | 1.0088 | 0.9894 |
| | $w_1/w_2/w_3$ = 0.9563/0.0359/0.0078<br>($x_1/x_2/x_3$ = 0.350/0.542/0.108) | | | $w_1/w_2/w_3$ = 0.9792/0.0049/0.0159<br>($x_1/x_2/x_3$ = 0.548/0.113/0.339) | | |
| 0.1 | 0.9978 | 0.9825 | 0.9592 | 0.9944 | 0.9792 | 0.9560 |
| 10 | 1.0040 | 0.9892 | 0.9669 | 1.0006 | 0.9859 | 0.9638 |
| 15 | 1.0069 | 0.9925 | 0.9705 | 1.0036 | 0.9893 | 0.9675 |
| 20 | 1.0098 | 0.9956 | 0.9740 | 1.0065 | 0.9924 | 0.9709 |
| 25 | 1.0126 | 0.9986 | 0.9774 | 1.0093 | 0.9954 | 0.9744 |
| 30 | 1.0154 | 1.0016 | 0.9807 | 1.0121 | 0.9984 | 0.9776 |
| 35 | 1.0181 | 1.0044 | 0.9839 | 1.0147 | 1.0013 | 0.9809 |
| 40 | 1.0207 | 1.0072 | 0.9870 | 1.0173 | 1.0041 | 0.9840 |
| 45 | 1.0232 | 1.0099 | 0.9900 | 1.0198 | 1.0068 | 0.9870 |
| 50 | 1.0257 | 1.0125 | 0.9929 | 1.0223 | 1.0094 | 0.9898 |
| | $w_1/w_2/w_3$ = 0.9800/0.0147/0.0053<br>($x_1/x_2/x_3$ = 0.548/0.339/0.113) | | | | | |
| 0.1 | 0.9966 | 0.9812 | 0.9581 | | | |
| 10 | 1.0027 | 0.9881 | 0.9658 | | | |
| 15 | 1.0057 | 0.9913 | 0.9695 | | | |
| 20 | 1.0087 | 0.9945 | 0.9730 | | | |
| 25 | 1.0114 | 0.9975 | 0.9764 | | | |
| 30 | 1.0142 | 1.0005 | 0.9797 | | | |
| 35 | 1.0168 | 1.0033 | 0.9828 | | | |
| 40 | 1.0194 | 1.0061 | 0.9860 | | | |
| 45 | 1.0219 | 1.0088 | 0.9890 | | | |
| 50 | 1.0244 | 1.0114 | 0.9919 | | | |

By definition, the volume change of mixing or excess volume ($V^E$) can be calculated from experimental density data via

$$V^E = V_m - \sum_{i=1}^{c} x_i V_i^o \tag{5}$$

with

$$V_m = \sum_{i=1}^{c} x_i M_i/\rho \tag{6}$$

where $V_m$ is the molar volume of a mixture and $c$ is the number of components. $x_i$, $V_i^o$, and $M_i$ are the mol fraction, molar volume, and molecular weight, respectively, for component $i$. The uncertainty of the calculated excess volumes is estimated to be within ±0.02 cm$^3$·mol$^{-1}$. The excess volumes of 1-octanol + acetophenone are positive over the entire experimental conditions. While the excess volumes increase substantially with increasing temperature, the pressure effect is insignificant. Figure 1 shows the excess volumes at 0.1 MPa varying with composition for 1-octanol + acetophenone. The excess volumes at constant temperature and pressure were correlated with a Redlich−Kister-type equation

$$V^E/x_1x_2 = \sum_{k=1}^{2} E_k(x_1 - x_2)^{k-1} \tag{7}$$

The optimial values of $E_k$ were determined by minimization of the following objective function ($\pi_2$)

$$\pi_2 = [\sum_{k=1}^{n} |V^E_{k,\mathrm{calc}} - V^E_{k,\mathrm{exp}}|]/n \tag{8}$$

The correlated results are listed in Table 3 and represented by the dashed curves in Figure 1.

***Ternary Systems.*** The *P*−*V*−*T* properties were also measured, over the same temperature and pressure ranges, for PPG-4000 with mixed solvents of 1-octanol and ac-

**Table 5. *P*−*V*−*T* Properties of PEG-200 (1) + PEGME-350 (2) + Anisole (3)**

| | $\rho$/g·cm$^{-3}$ | | | $\rho$/g·cm$^{-3}$ | | |
|---|---|---|---|---|---|---|
| *P*/MPa | 298.15 K | 318.15 K | 348.15 K | 298.15 K | 318.15 K | 348.15 K |
| | $w_1/w_2/w_3 = 0.1773/0.7489/0.0738$<br>$(x_1/x_2/x_3 = 0.200/0.600/0.200)$ | | | $w_1/w_2/w_3 = 0.2736/0.3851/0.3413$<br>$(x_1/x_2/x_3 = 0.200/0.200/0.600)$ | | |
| 0.1 | 1.0868 | 1.0700 | 1.0433 | 1.0687 | 1.0510 | 1.0246 |
| 10 | 1.0913 | 1.0752 | 1.0493 | 1.0740 | 1.0570 | 1.0313 |
| 15 | 1.0936 | 1.0777 | 1.0523 | 1.0766 | 1.0598 | 1.0346 |
| 20 | 1.0959 | 1.0801 | 1.0551 | 1.0791 | 1.0625 | 1.0377 |
| 25 | 1.0981 | 1.0825 | 1.0577 | 1.0816 | 1.0651 | 1.0408 |
| 30 | 1.1002 | 1.0847 | 1.0604 | 1.0840 | 1.0678 | 1.0437 |
| 35 | 1.1023 | 1.0870 | 1.0629 | 1.0864 | 1.0704 | 1.0466 |
| 40 | 1.1043 | 1.0892 | 1.0655 | 1.0886 | 1.0728 | 1.0494 |
| 45 | 1.1063 | 1.0914 | 1.0679 | 1.0909 | 1.0753 | 1.0521 |
| 50 | 1.1083 | 1.0935 | 1.0702 | 1.0932 | 1.0777 | 1.0548 |
| | $w_1/w_2/w_3 = 0.6219/0.2918/0.0862$<br>$(x_1/x_2/x_3 = 0.600/0.200/0.200)$ | | | $w_1/w_2/w_3 = 0.3713/0.4853/0.1434$<br>$(x_1/x_2/x_3 = 0.350/0.325/0.325)$ | | |
| 0.1 | 1.0989 | 1.0820 | 1.0572 | 1.0793 | 1.0622 | 1.0369 |
| 10 | 1.1035 | 1.0874 | 1.0631 | 1.0842 | 1.0678 | 1.0430 |
| 15 | 1.1058 | 1.0899 | 1.0659 | 1.0866 | 1.0703 | 1.0460 |
| 20 | 1.1081 | 1.0924 | 1.0687 | 1.0889 | 1.0729 | 1.0490 |
| 25 | 1.1103 | 1.0949 | 1.0714 | 1.0912 | 1.0755 | 1.0517 |
| 30 | 1.1125 | 1.0972 | 1.0740 | 1.0935 | 1.0779 | 1.0545 |
| 35 | 1.1146 | 1.0995 | 1.0766 | 1.0957 | 1.0803 | 1.0571 |
| 40 | 1.1167 | 1.1017 | 1.0791 | 1.0978 | 1.0826 | 1.0597 |
| 45 | 1.1188 | 1.1040 | 1.0816 | 1.0999 | 1.0849 | 1.0623 |
| 50 | 1.1209 | 1.1062 | 1.0839 | 1.1020 | 1.0872 | 1.0647 |

**Table 6. Results of Specific-Volume Correlation with the Equations of State for “Pure” Compounds**

| | FOV EOS | | | | Schotte EOS | | | |
|---|---|---|---|---|---|---|---|---|
| | $P^*$ | $T^*$ | $V^*$ | $\hat{V}$ AAD$^a$ | $P^*$ | $T^*$ | $V^*$ | $\hat{V}$ AAD$^a$ |
| compound | MPa | K | cm$^3$·g$^{-1}$ | % | MPa | K | cm$^3$·g$^{-1}$ | % |
| 1-octanol | 494.9 | 5627 | 0.9947 | 0.05 | 511.1 | 4945 | 0.9794 | 0.04 |
| acetophenone | 627.4 | 5838 | 0.8076 | 0.03 | 649.2 | 5138 | 0.7954 | 0.02 |
| anisole | 640.2 | 5363 | 0.8164 | 0.02 | 651.3 | 4753 | 0.8058 | 0.03 |
| PPG-4000 | 483.7 | 6227 | 0.8401 | 0.04 | 498.9 | 5543 | 0.8297 | 0.03 |
| PEG-200 | 725.1 | 6486 | 0.7551 | 0.03 | 767.4 | 5742 | 0.7449 | 0.02 |
| PEGME−350 | 686.2 | 6101 | 0.7695 | 0.02 | 702.1 | 5393 | 0.7589 | 0.02 |

[a] $\hat{V}\ \mathrm{AAD} = \frac{100}{n}\sum_{k=1}^{n} |\hat{V}_{k,\mathrm{calc}} - \hat{V}_{k,\mathrm{exp}}|/\hat{V}_{k,\mathrm{exp}}$.

**Table 7. Results of Specific-Volume Correlation with the Equations of State for Polymer Solutions**

| | FOV EOS | | Schotte EOS | |
|---|---|---|---|---|
| mixture (1) + (2) | $\Delta_{12}$ | $\hat{V}$ AAD$^a$/% | $\Delta_{12}$ | $\hat{V}$ AAD$^a$/% |
| 1-octanol + acetophenone | 0.0162 | 0.06 | 0.0152 | 0.06 |
| PPG-4000 + 1-octanol$^b$ | 0.0002 | 0.06 | −0.0004 | 0.06 |
| PPG-4000 + acetophenone$^b$ | −0.0430 | 0.06 | −0.0439 | 0.05 |
| PEG-200 + anisole$^c$ | −0.0299 | 0.15 | −0.0317 | 0.14 |
| PEGME-350 + anisole$^c$ | −0.1294 | 0.22 | −0.1290 | 0.21 |
| PEG-200 + PEGME-350$^d$ | −0.0020 | 0.02 | −0.0012 | 0.01 |

[a] $\hat{V}\ \mathrm{AAD} = \frac{100}{n}\sum_{k=1}^{n} |\hat{V}_{k,\mathrm{calc}} - \hat{V}_{k,\mathrm{exp}}|/\hat{V}_{k,\mathrm{exp}}$. [b] Data taken from Lin et al.[13] [c] Data taken from Lee et al.[12] [d] Data taken from Lee et al.[9]

etophenone and for the polymer blends of PEG-200/PEGME-350 with anisole. The experimental densities are tabulated in Tables 4 and 5 for PPG + 1-octanol + acetophenone and PEG + PEGME + anisole, respectively. These volumetric data form a basis to test the capability of polymer equations of state for multicomponent polymer solutions.

## *P*−*V*−*T* Property Calculations with Equations of State

Two polymer EOSs, the FOV[18] and the Schotte,[19] were tested in the present study with the experimental volumetric properties. The definitions of these two EOSs are as follows.

The FOV EOS

$$\frac{\overline{P}\overline{V}}{\overline{T}} = \frac{\overline{V}^{1/3}}{\overline{V}^{1/3} - 1} - \frac{1}{\overline{T}\overline{V}} \tag{9}$$

The Schotte EOS

$$\frac{\overline{P}\overline{V}}{\overline{T}} = \frac{RT^*}{P^*MV^*}\left(1 - \frac{1}{\overline{V}^{1/3}}\right) + \frac{1}{\overline{V}^{1/3} - 1} - \frac{1}{\overline{T}\overline{V}} \tag{10}$$

where $M$ is the molecular weight, $\bar{P} = P/P^*$, $\bar{V} = V/V^*$, and $\bar{T} = T/T^*$. The model parameters $P^*$, $V^*$, and $T^*$ are characteristic pressure, specific volume, and temperature,

**Table 8. Predicted Results from the FOV and the Schotte EOSs for the Ternary Polymer Solutions**

| PEG-4000 (1) + acetophenone (2) + 1-octanol (3) | | | PEG-200 (1) + PEGME−350 (2) + anisole (3) | | |
|---|---|---|---|---|---|
| | FOV | Schotte | | FOV | Schotte |
| $x_1/x_2/x_3$ | $\hat{V}$ AAD$^a$/% | $\hat{V}$ AAD$^a$/% | $x_1/x_2/x_3$ | $\hat{V}$ AAD$^a$/% | $\hat{V}$ AAD$^a$/% |
| 0.168/0.312/0.520 | 0.06 | 0.05 | 0.2/0.6/0.2 | 0.12 | 0.10 |
| 0.168/0.520/0.312 | 0.06 | 0.06 | 0.2/0.2/0.6 | 0.13 | 0.12 |
| 0.350/0.108/0.542 | 0.06 | 0.04 | 0.6/0.2/0.2 | 0.12 | 0.11 |
| 0.350/0.325/0.325 | 0.05 | 0.04 | 0.35/0.325/0.325 | 0.12 | 0.13 |
| 0.350/0.542/0.108 | 0.06 | 0.05 | | | |
| 0.548/0.113/0.339 | 0.06 | 0.05 | | | |
| 0.548/0.339/0.113 | 0.09 | 0.09 | | | |

[a] $\hat{V}\ \mathrm{AAD} = \dfrac{100}{n}\sum_{k=1}^{n} |\hat{V}_{k,\mathrm{calc}} - \hat{V}_{k,\mathrm{exp}}|/\hat{V}_{k,\mathrm{exp}}$.

respectively, which were determined by fitting the EOS to experimental $P-V-T$ data for each constituent. The characteristic parameters of 1-octanol, acetophenone, anisole, PPG-4000, PEG-200, and PEGME-350 are given in Table 6. The mixing rules proposed by Schotte[19] were used in this study

$$V_{\mathrm{m}}^{*} = \left[M_{\mathrm{m}}\left(\sum_{i=1}^{c} \frac{\Psi_i}{\mathrm{M}_i V_i^{*}}\right)\right]^{-1} \tag{11}$$

$$T_m^{*} = \frac{P_m^{*}}{\displaystyle\sum_{i=1}^{c} \frac{\Psi_i P_i^{*}}{T_i^{*}}} \tag{12}$$

and

$$P_m^{*} = \sum_{i=1}^{c}\sum_{j=1}^{c} \Psi_i \Psi_j P_{ij}^{*} \tag{13}$$

with

$$\Psi_i = \frac{w_i V_i^{*}}{\displaystyle\sum_{i=1}^{c} w_i V_i^{*}} \tag{14}$$

and

$$P_{ij}^{*} = (1 - \Delta_{ij})(P_i^{*} P_j^{*})^{0.5} \tag{15}$$

where $\Psi_i$, $M_i$, and $w_i$ refer to the segment volume fraction, the number average molecular weight, and the weight fraction of component $i$, respectively. The parameter $\Delta_{ij}$ in eq 15 is a binary interaction constant for the $i-j$ pair. Its value was determined from the $P-V-T$ data of each binary system by minimization of the absolute average deviation (AAD) of the calculated specific volume. Table 7 reports the correlated results. Both the FOV and the Schotte EOS calculate quantitatively the specific volumes for the constituent binaries. With these optimized $\Delta_{ij}$ values, the FOV and the Schotte EOSs were employed to predict the volumetric properties for mixtures of PPG with mixed solvents of 1-octanol + acetophenone and the polymer blends of PEG/PEGME with anisole. The predicted results are presented in Table 8. The AADs of predicted specific volumes are better than 0.13% from both EOSs, and these two EOSs are almost equally well for the investigated ternary systems. Figures 2 and 3 compare the predicted densities with the experimental values for PPG + 1-octanol + acetophenone and PEG + PEGME + anisole, respectively. The agreement is satisfactory.

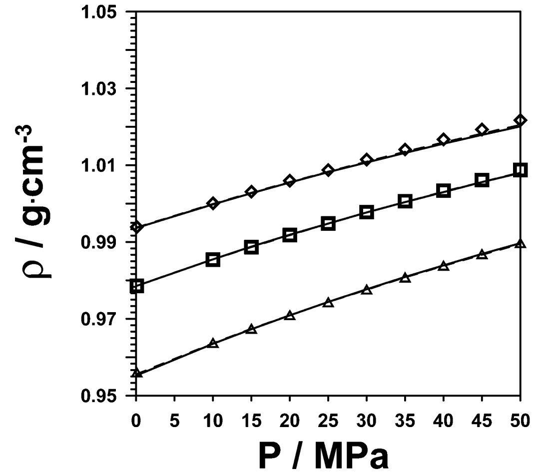

**Figure 2.** Comparison of predicted results with experimental values for PPG-4000 (1) + acetophenone (2) + 1-octanol (3) of $x_1/x_2 = 0.35/0.325$ at ◇, 298.15 K; □, 318.15 K, △, 348.15 K; solid line, calculated from the FOV EOS; dashed line, calculated from the Schotte EOS.

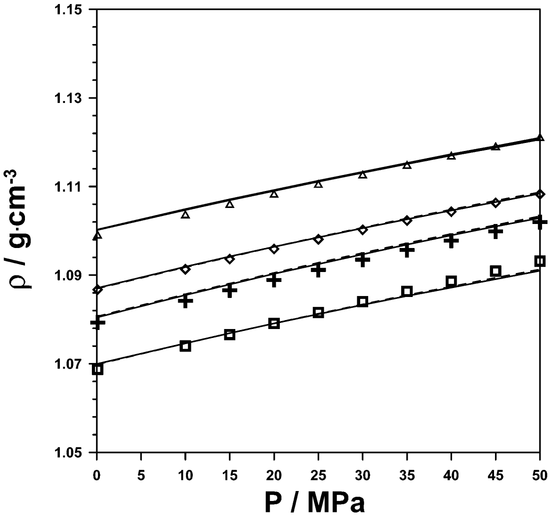

**Figure 3.** Comparison of predicted results with experimental values for PEG-200 (1) + PEGME-350 (2) + anisole (3) at 298.15 K: ◇, $x_1/x_2 = 0.2/0.6$; □, $x_1/x_2 = 0.2/0.2$; △, $x_1/x_2 = 0.6/0.2$; +, $x_1/x_2 = 0.35/0.325$; solid line, calculated from the FOV EOS; dashed line, calculated from the Schotte EOS.

## Conclusions

The properties of $P-V-T$ have been measured for 1-octanol + acetophenone, PPG-4000 + 1-octanol + acetophenone, and PEG-200 + PEGME-350 + anisole at temperatures from 298.15 K to 348.15 K and pressures up to 50 MPa. The Tait equation represented accurately the pressure effect on liquid density for 1-octanol + acetophenone. The excess volumes are positive for 1-octanol + acetophenone over the entire composition range. It was also found that the excess volumes increased with an increase of temperature but were insensitive to pressure. Both the FOV and the Schotte EOSs correlated accurately the $P-V-T$ data of the constituent binaries. Moreover, these two polymer EOSs predicted the specific volumes to an AAD better than 0.13% for the polymer solutions of PPG with the mixed solvent of 1-octanol/acetophenon and the polymer blends of PEG/PEGME with anisole, over the entire experimental conditions.

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