

Effect of Aqueous Solution of Tri-potassium Citrate on the Volumetric Behavior of Poly(propylene glycol) 400 at $T = (288.15 \text{ to } 313.15) \text{ K}$

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Density and ultrasonic velocity of poly(propylene glycol) in aqueous solutions of tri-potassium citrate have been measured at $T = (288.15 \text{ to } 313.15) \text{ K}$ at atmospheric pressure. From these measurements, the excess specific volume, isentropic compressibility, and isentropic compressibility deviation values have been determined. It has been found that the excess specific volume and isentropic compressibility deviation are negative and decrease in magnitude as temperature or concentration of tri-potassium citrate increases.

Introduction

Ternary aqueous solutions of poly(propylene glycol) (PPG) and certain electrolytes separate into a PPG-rich and a salt-rich phase over part of the composition space. Aqueous two-phase systems of this nature are useful for the separation and purification of biomaterials where the employment of a nonpolar solvent/aqueous phase system would lead to degradation of biological activity.^{1,2} Citrate is biodegradable and nontoxic and could be discharged into biological wastewater treatment plants; therefore, the citrates can be considered as a substitute for inorganic salts because the citrate forms aqueous two-phase system with PPG that are suitable for protein extraction. Liquid–liquid equilibrium data for some aqueous PPG + salt two-phase systems have been reported in the literature.^{3–7} Thermodynamic properties of aqueous polymer–salt systems are necessary for a fundamental understanding of the phase-forming ability and also for the development of theoretical models for the prediction of the partitioning behavior of the two-phase system. Although knowledge of acoustical and volumetric properties of aqueous polymer–salt solutions provides useful information on the interactions existing in these systems, only a limited amount of experimental work has been carried out investigating the volumetric properties of PPG + salt + water systems, and existing data are limited to a single temperature $T = 298.15 \text{ K}$.^{5,6,8}

The present work presents experimental data on the density and ultrasonic velocity of PPG in aqueous solutions of 0.0206 and 0.0397 mass fraction tri-potassium citrate (K_3Cit) at $T = (288.15 \text{ to } 313.15) \text{ K}$. The values of the excess specific volume and isentropic compressibility were then calculated from the measured data. Although there are some reports on the density and ultrasonic velocity of binary aqueous PPG solutions,^{9–13} there are no literature data on the density and ultrasonic velocity of aqueous PPG + K_3Cit solutions, which is a very important system in respect to its use in biotechnology.

Experimental Section

Materials. PPG with a quoted molar mass of $400 \text{ g}\cdot\text{mol}^{-1}$ was obtained from Fluka. The manufacturer has characterized this polymer as P400 with lot and filling code 1252304 20206282. Potassium citrate ($\text{C}_6\text{H}_5\text{K}_3\text{O}_7\cdot\text{H}_2\text{O}$) with a minimum

purity of 99 % was obtained from Fluka. The polymer and salt were used without further purification, and double-distilled, deionized water was used.

Apparatus and Procedures. All the solutions were prepared by mass on a Sartorius CP124S balance precise to within $\pm 0.0001 \text{ g}$. The density and sound velocity of the mixtures were measured at different temperatures with a digital vibrating-tube analyzer (Anton Paar DSA 5000, Austria) with proportional temperature control that kept the samples at working temperature within $\pm 10^{-3} \text{ K}$. The apparatus was calibrated with double-distilled, deionized, and degassed water and dry air at atmospheric pressure according to the instrument catalog. Densities and ultrasonic velocities can be measured to $\pm 10^{-6} \text{ g}\cdot\text{cm}^{-3}$ and $\pm 10^{-2} \text{ m}\cdot\text{s}^{-1}$, respectively, under the most favorable conditions. The uncertainties of density and ultrasonic velocity measurements were $\pm 3 \times 10^{-6} \text{ g}\cdot\text{cm}^{-3}$ and $\pm 10^{-1} \text{ m}\cdot\text{s}^{-1}$, respectively.

Results and Discussion

Experimental data of density and ultrasonic velocity for various PPG + K_3Cit + H_2O solutions determined at $T = (288.15, 293.15, 298.15, 303.15, 308.15, \text{ and } 313.15) \text{ K}$ are given in Table 1. Repeat determinations of the solution densities and ultrasonic velocities were well within the maximum allowed error. The values obtained for the density of water, as a function of temperature, were in agreement with accepted values. That is to say, the two last solutions of PPG in aqueous solution of mass fraction 0.0397 K_3Cit at 313.15 K were two-phase systems; therefore, density and ultrasonic velocity data were not reported in Table 1.

The excess specific volumes (V^E) of the solution were calculated using the relation:

$$V^E = \frac{1}{d} - \frac{w_p}{d_p} - \frac{(1 - w_p)}{d_s} \quad (1)$$

where w_p , d , d_p , and d_s are polymer mass fraction, density of solution, density of pure polymer, and density of solvent (tri-potassium citrate + water is considered as the solvent), respectively. In Figures 1 and 2, the temperature and concentration dependence of V^E are shown. From the Figures 1 and 2, it can be seen that the V^E values are negative and become more negative as temperature decreases. The similar behavior has been

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Table 1. Density d and Ultrasonic Velocity u of PPG in Aqueous K_3Cit Solutions as a Function of the Polymer Mass Fraction w_p and Temperature T

w_p	$T/K = 288.15$ K		$T/K = 293.15$ K		$T/K = 298.15$ K		$T/K = 303.15$ K		$T/K = 308.15$ K		$T/K = 313.15$ K	
	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$	d $g \cdot cm^{-3}$	u $m \cdot s^{-1}$
In Aqueous Solution of Mass Fraction 0.0206 K_3Cit												
0.0000	1.012325	1480.79	1.011318	1496.34	1.010065	1510.11	1.008592	1522.11	1.006912	1532.44	1.005041	1541.22
0.0015	1.012410	1481.69	1.011401	1497.21	1.010142	1510.89	1.008665	1522.80	1.006982	1533.06	1.005108	1541.82
0.0030	1.012525	1482.78	1.011511	1498.11	1.010250	1511.73	1.008768	1523.60	1.007079	1533.78	1.005204	1542.42
0.0060	1.012761	1485.00	1.011738	1500.25	1.010469	1513.68	1.008977	1525.38	1.007282	1535.41	1.005396	1543.89
0.0100	1.013056	1487.93	1.012021	1502.98	1.010740	1516.21	1.009239	1527.73	1.007534	1537.59	1.005640	1545.93
0.0199	1.013840	1494.92	1.012773	1509.25	1.011464	1521.98	1.009933	1532.97	1.008201	1542.33	1.006279	1550.16
0.0301	1.014649	1502.11	1.013549	1515.90	1.012207	1528.06	1.010647	1538.51	1.008885	1547.35	1.006936	1554.71
0.0401	1.015454	1509.21	1.014318	1522.48	1.012944	1534.03	1.011351	1543.91	1.009560	1552.22	1.007578	1559.09
0.0601	1.017086	1523.41	1.015874	1535.30	1.014429	1545.64	1.012766	1554.42	1.010905	1561.67	1.008859	1567.49
0.0800	1.018760	1537.22	1.017465	1547.88	1.015937	1556.99	1.014198	1564.53	1.012262	1570.57	1.010142	1575.20
0.0998	1.020450	1550.88	1.019064	1560.16	1.017447	1567.90	1.015623	1574.12	1.013602	1578.88	1.011398	1582.22
0.1297	1.023049	1570.91	1.021510	1577.86	1.019743	1583.40	1.017770	1587.46	1.015604	1590.06	1.013253	1591.16
0.1583	1.025580	1589.17	1.023869	1593.79	1.021940	1596.98	1.019807	1598.70	1.017480	1598.87	1.014959	1597.37
0.1942	1.028781	1610.25	1.026828	1611.59	1.024660	1611.51	1.022290	1609.90	1.019723	1606.57	1.016948	1601.10
0.2468	1.033295	1635.08	1.030929	1631.23	1.028357	1625.85	1.025586	1618.80	1.022607	1609.69	1.019381	1597.70
0.2885	1.036614	1647.94	1.033887	1639.87	1.030960	1630.28	1.027838	1618.95	1.024498	1605.48	1.020859	1589.03
0.3482	1.040701	1654.00	1.037444	1640.83	1.034007	1626.31	1.030382	1610.42	1.026538	1592.63	1.022168	1573.07
In Aqueous Solution of Mass Fraction 0.0397 K_3Cit												
0.0000	1.024697	1493.33	1.023594	1508.47	1.022258	1521.87	1.020714	1533.50	1.018978	1543.49	1.017060	1551.96
0.0015	1.024769	1494.41	1.023664	1509.33	1.022326	1522.67	1.020778	1534.26	1.019035	1544.19	1.017114	1552.59
0.0030	1.024871	1495.43	1.02376	1510.38	1.022416	1523.64	1.020866	1535.11	1.019122	1544.96	1.017195	1553.30
0.0061	1.025076	1497.66	1.023958	1512.36	1.022607	1525.45	1.021048	1536.78	1.019293	1546.49	1.017363	1554.68
0.0101	1.025324	1500.43	1.024192	1514.99	1.022832	1527.86	1.021260	1538.97	1.019498	1548.47	1.017554	1556.49
0.0200	1.025990	1507.57	1.024829	1521.58	1.023438	1533.87	1.021841	1544.44	1.020053	1553.46	1.018082	1561.00
0.0299	1.026654	1514.68	1.025461	1528.05	1.024041	1539.78	1.022415	1549.87	1.020595	1558.40	1.01860	1565.49
0.0403	1.027369	1521.97	1.026139	1534.73	1.024685	1545.83	1.023025	1555.31	1.021174	1563.26	1.019145	1569.77
0.0602	1.028758	1535.86	1.027455	1547.31	1.025930	1557.23	1.024201	1565.54	1.022281	1572.36	1.020186	1577.76
0.0800	1.030186	1549.45	1.028801	1559.52	1.027198	1568.12	1.025389	1575.20	1.023394	1580.78	1.021220	1584.96
0.1010	1.031725	1563.56	1.030242	1572.18	1.028542	1579.31	1.026642	1584.90	1.024553	1589.02	1.022284	1591.69
0.1300	1.033895	1582.35	1.032261	1588.69	1.030410	1593.55	1.028363	1596.89	1.026121	1598.66	1.023692	1598.70
0.1588	1.036056	1599.77	1.034248	1603.61	1.032225	1605.94	1.030020	1606.67	1.027583	1605.61	1.024942	1602.24
0.2006	1.039174	1621.96	1.037069	1621.75	1.034754	1619.90	1.032231	1616.18	1.02949	1610.08	1.025956	1599.10
0.2522	1.042799	1641.95	1.040269	1636.25	1.037522	1628.60	1.034556	1618.68	1.031311	1605.67	1.026520	1588.72
0.2983	1.045658	1650.98	1.042702	1640.39	1.039534	1627.81	1.036126	1612.95	1.032294	1595.01		
0.3466	1.048027	1651.72	1.044632	1637.57	1.041029	1621.58	1.037165	1603.73	1.032513	1585.45		

observed for the binary aqueous PPG solutions.¹³ Two samples of the same polymer (e.g., equal in average molecular weight) may exhibit different physicochemical properties if they differ in their molecular weight distribution. However, the polymer used in this work has a small molar mass; therefore, the

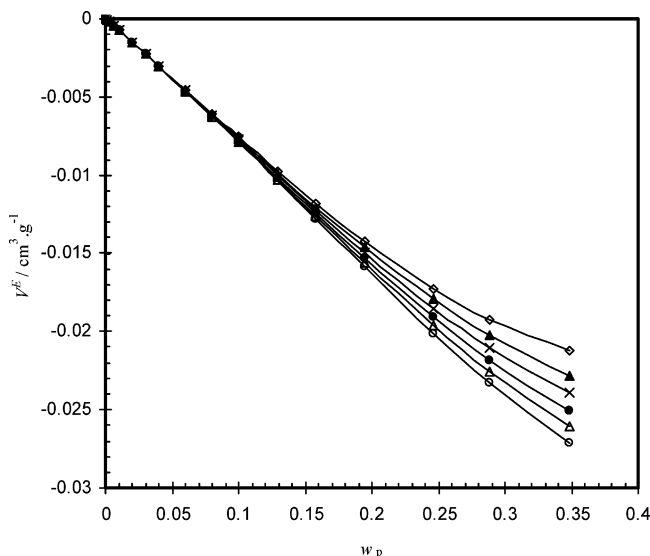


Figure 1. Excess specific volumes V^E of PPG in aqueous solution of mass fraction 0.0206 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: \circ , $T = 288.15$ K; \triangle , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

polydispersity of this polymer is small. In fact, we obtained several density and ultrasonic velocity data for binary aqueous PPG solution and from which the excess volume and isentropic compressibility were determined and compared with those of ref 13. We found that there is a very good agreement between our data and those taken from ref 13.

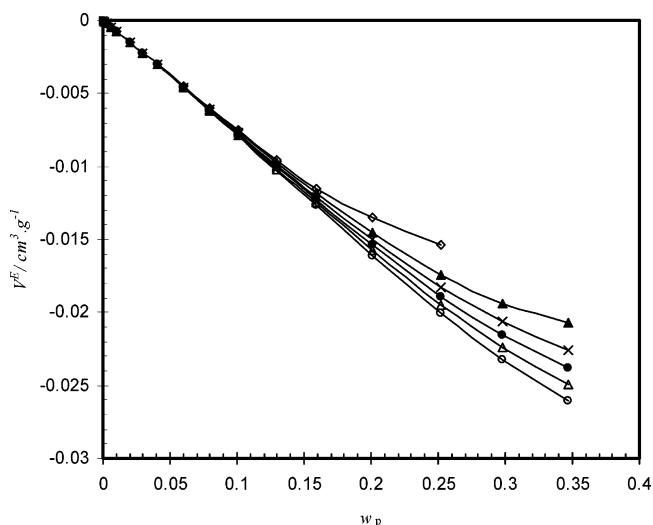


Figure 2. Excess specific volumes V^E of PPG in aqueous solution of mass fraction 0.0397 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: \circ , $T = 288.15$ K; \triangle , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

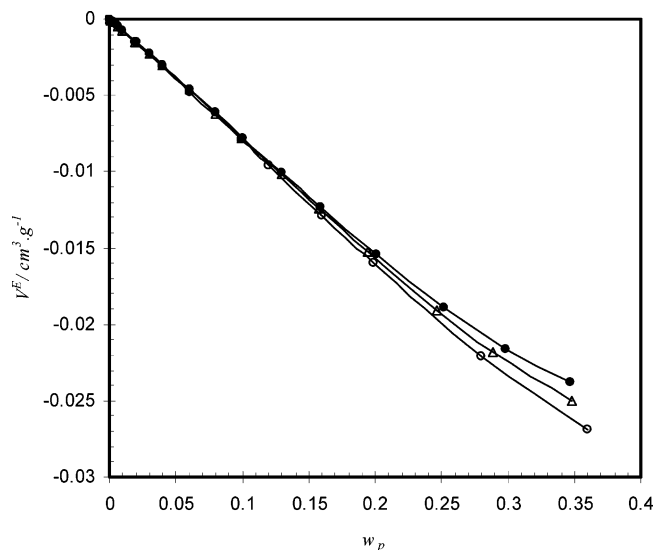


Figure 3. Excess specific volumes V^E of PPG solutions plotted against mass fraction of polymer w_p at $T = 298.15$ K: \circ , in water; Δ , in aqueous solution of mass fraction 0.0206 K_3Cit ; \bullet , in aqueous solution of mass fraction 0.0397 K_3Cit .

Mainly, the behavior of V^E is attributed to the breakdown of the H_2O self-associated molecules from each other (a positive volume), the breakdown of the PPG self-associated molecules from each other (a positive volume), the breakdown of the ion-dipole interaction of H_2O molecules with the salt (a positive volume), the repulsive interactions between the anion and the anionic-like polyether group of PPG (a positive volume), and the negative contribution of volume due to the intermolecular interactions between the hydrogen atom of the water and the oxygen atom of the PPG and difference between the size of water and polymer (the relatively small water molecules and ions fit in the available free volume of polymer upon mixing). The negative values of V^E show that effects due to the hydrogen bond interactions between PPG and water and packing effect are predominant factors. At higher temperatures, hydrogen bond interactions are weakened; hence, less negative values of the excess specific volumes are obtained. Figure 3 shows that the excess specific volume becomes less negative as salt concentration increases. As mentioned above, ternary aqueous solution of PPG and tri-potassium citrate separates into a PPG-rich phase and a salt-rich phase over part of the composition space. The formation of aqueous two-phase systems clearly indicates the mutual exclusion of the ions and the polymer and their high affinity for the solvent. It is possible that even in homogeneous systems (below the phase boundary) the ions are excluded from the near-surface region of the polymer in solution. With increase in the concentration of the polymer or the salt, the extent of exclusion will increase. Ultimately, the system could reach a state where, for entropic reasons, phase formation would become favorable. Exclusion of ions from the polymer molecule-water interface itself can occur for a number of reasons. Both the polymer and the ion are strongly hydrated in solution. Because of the hydration sheath, the near-surface region of the polymer may not be accessible to structure-making ions. Exclusion can occur also by repulsive interaction between the anions and the anionic-like polyether group of PPG especially in the presence of nonbonding cations like K^+ , NH_4^+ , and Na^+ . Ananthapadmanabhan and Goddard,¹⁴ using the turbidimetric titration technique, have shown that, for a series of sodium salts, two-phase formation occurs with anions such as sulfate, which have a marked salting-out effect on nonelectrolytes, but that for large,

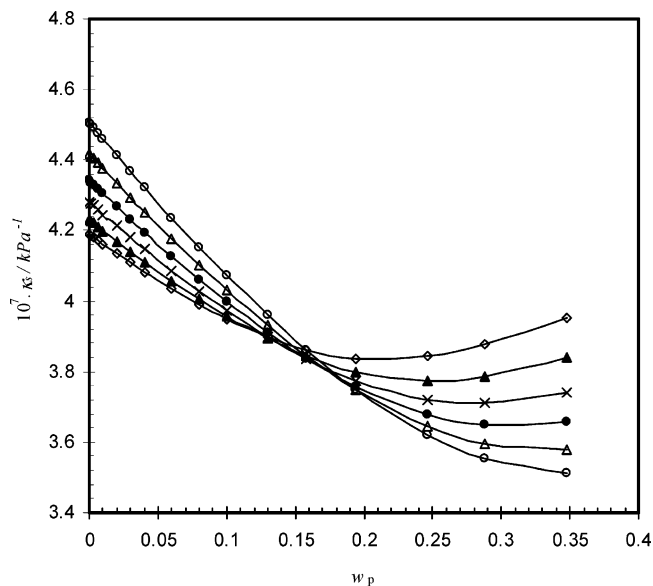


Figure 4. Isentropic compressibility κ_s of PPG in aqueous solution of mass fraction 0.0206 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: \circ , $T = 288.15$ K; Δ , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

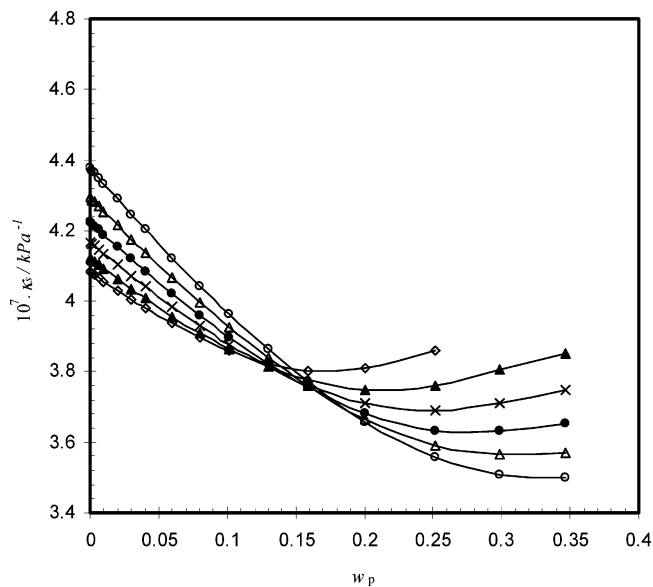


Figure 5. Isentropic compressibility κ_s of PPG in aqueous solution of mass fraction 0.0397 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: \circ , $T = 288.15$ K; Δ , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

singly charged ions such as iodide only one phase exists. The same authors also found that, for a series of sulfates, the salting-out effects of the anions are offset by di- and trivalent cations, which are believed to coordinate to the ether oxygens to produce a salting-in effect. The less negative V^E values for PPG in higher concentration of K_3Cit solutions are attributed to the repulsive interactions between the triply charged citrate anion and the anionic-like polyether group of PPG and also the ionic species that hydrate strongly evidently to induce the dehydration of PG units.

At different polymer mass fraction (w_p), the isentropic compressibility of solution (κ_s) determined by means of the Laplace equation ($\kappa_s = d^{-1}u^{-2}$) is shown at working temperatures in Figures 4 and 5. It was found that the compressibility of the investigated binary aqueous K_3Cit solutions and all ternary aqueous PPG + K_3Cit solutions at the working temperatures is

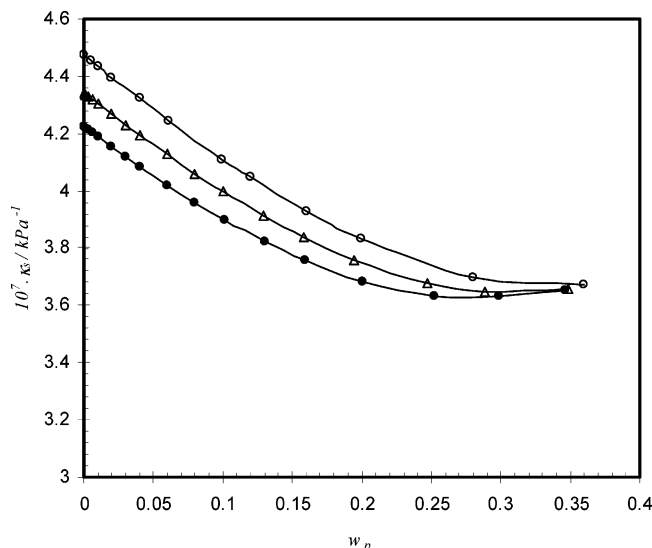


Figure 6. Isentropic compressibility κ_s of PPG solutions plotted against mass fraction of polymer w_p at $T = 298.15$ K: O, in water; Δ , in aqueous solution of mass fraction 0.0206 K_3Cit ; \bullet , in aqueous solution of mass fraction 0.0397 K_3Cit .

less than that of the pure PPG. This is due to the chain-like structure of the polymer and its greater free volume. Also the obtained data for pure solvent ($w_p = 0$) and pure PPG show that difference between compressibility of solvent and pure PPG becomes bigger at higher temperatures. From Figures 4 and 5, we note that the values of κ_s show a shallow minimum. This minimum becomes more distinct and shows a tendency to shift to a higher PPG concentration as the temperature is lowered. For low concentration of PPG, the values of κ_s decrease with the increase in the concentration of PPG, which is completely opposite that for high PPG concentrations. In fact in aqueous polymer solutions the isentropic compressibility is the sum of two contributions, $\kappa_s(\text{solvent intrinsic})$ and $\kappa_s(\text{solute intrinsic})$. Here $\kappa_s(\text{solvent intrinsic})$ is the isentropic compressibility due to the compression of the three-dimensional network structure of water and $\kappa_s(\text{solute intrinsic})$ is the isentropic compressibility due to the compression of the hydration shell of polymer segments, intermolecular distance of polymer chain (free volume), and incorporation of water molecules into the polymer chain. Figures 4 and 5 show that for the binary aqueous K_3Cit solution and ternary solutions with low concentration of PPG the measured values of κ_s decrease with the increase in temperature which is completely opposite that for high PPG concentrations and pure PPG. From Figures 4 and 5, we conclude that the $\kappa_s(\text{solvent intrinsic})$ is the dominant contribution to the total value of κ_s from pure solvent up to the converging concentration and beyond that $\kappa_s(\text{solute intrinsic})$ is the substantial contribution. From Figures 4 and 5, it can be seen that the compressibility isotherms for all the measured temperatures intersect approximately at a fixed polymer mass fraction close to 0.15. Therefore, one can assume that the isentropic compressibilities for the corresponding concentration are independent of temperature and at this concentration $d\kappa_s(\text{solute intrinsic})/dT + d\kappa_s(\text{solvent intrinsic})/dT = 0$.

In Figure 6, the values of κ_s are plotted as a function of PPG mass fraction at various salt concentration at 298.15 K. Figure 6 shows that, at 298.15 K at a constant polymer concentration, the values of κ_s decreases as the concentration of salt increases and also the minimum in the compressibility isotherms shows a tendency to shift to a lower PPG concentration as the concentration of salt is increased. In fact, as can be seen from

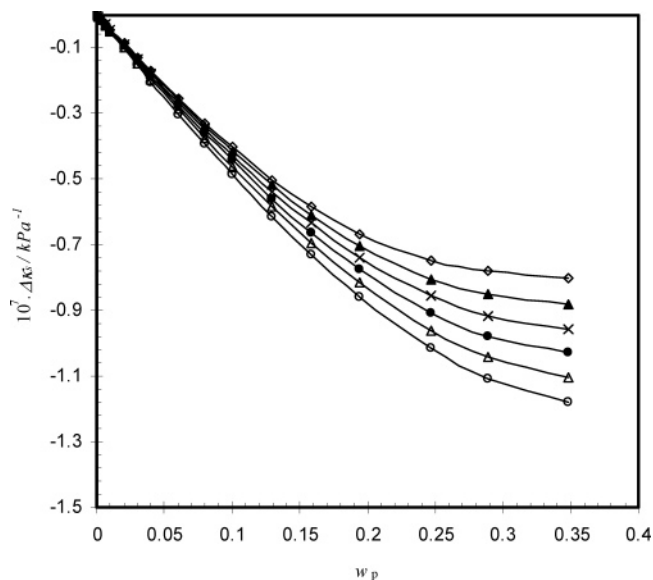


Figure 7. Isentropic compressibility increments $\Delta\kappa_s$ of PPG in aqueous solution of mass fraction 0.0206 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: O, $T = 288.15$ K; Δ , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

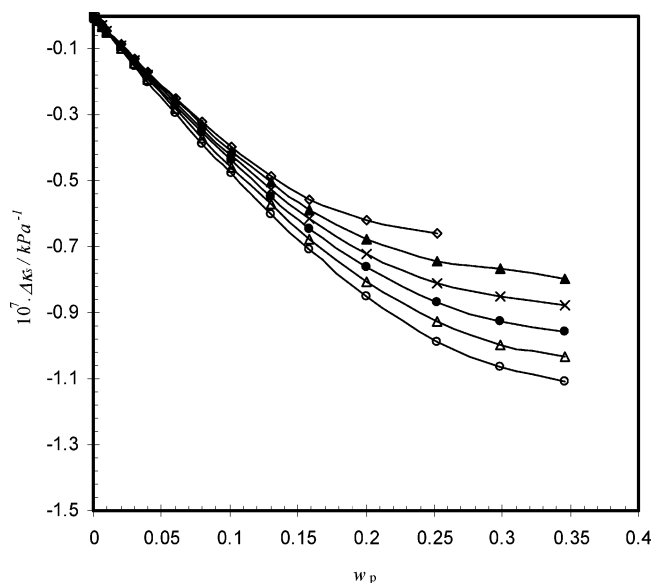


Figure 8. Isentropic compressibility increments $\Delta\kappa_s$ of PPG in aqueous solution of mass fraction 0.0397 K_3Cit plotted against mass fraction of polymer w_p at different temperatures: O, $T = 288.15$ K; Δ , $T = 293.15$ K; \bullet , $T = 298.15$ K; \times , $T = 303.15$ K; \blacktriangle , $T = 308.15$ K; \diamond , $T = 313.15$ K.

Figure 6 at a constant polymer concentration, isentropic compressibility of a solution of PPG in water is larger than that of a solution of PPG in water + K_3Cit . This effect can be attributed to the strong attractive interactions due to the hydration of ions and also incorporation of ionic species into the PPG chain. Figure 6 shows that, at 298.15 K as the concentration of tri-potassium citrate is increased, κ_s of water + K_3Cit solution is decreased due to the combined effect of hydration of ions and breaking of three-dimensional network structure of water. The similar behavior is observed for the other temperatures.

The experimental isentropic compressibility increments ($\Delta\kappa_s$) are obtained using the relation:

$$\Delta\kappa_s = \kappa_s - w_p\kappa_{s,p} - (1 - w_p)\kappa_{s,s} \quad (2)$$

where $\kappa_{s,p}$ and $\kappa_{s,s}$ are the values of isentropic compressibility

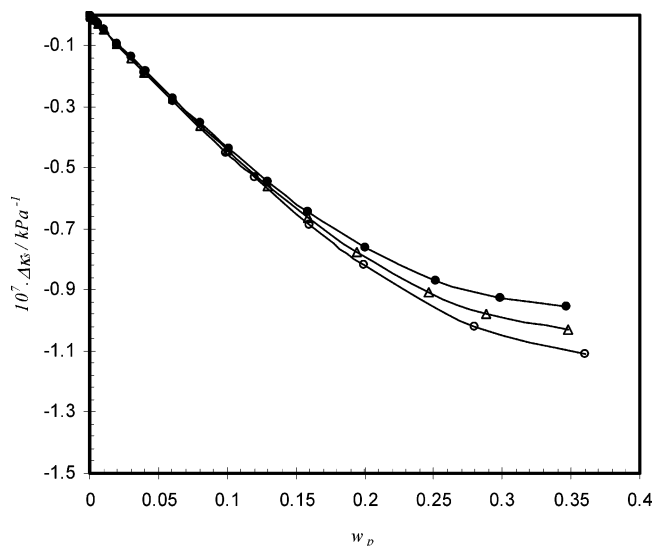


Figure 9. Isentropic compressibility increments $\Delta\kappa_s$ of PPG solutions plotted against mass fraction of polymer w_p at $T = 298.15$ K: O, in water; Δ , in aqueous solution of mass fraction 0.0206 K_3Cit ; \bullet , in aqueous solution of mass fraction 0.0397 K_3Cit .

of pure polymer and solvent (tri-potassium citrate + water is considered as the solvent), respectively. In Figures 7 and 8, the temperature and concentration dependence of $\Delta\kappa_s$ have been given. From Figures 7 and 8, it can be seen that the $\Delta\kappa_s$ values are negative and become more negative as temperature decreases. The similar behavior has been observed for the binary aqueous PPG solutions.¹³ The behavior of $\Delta\kappa_s$ implies a great difficulty in compressing the components of the mixture regarding those that can be expected under an ideal behavior. This is due to a closer approach of unlike molecules and stronger interaction between components of mixtures that lead to decreasing compressibility.

In Figure 9, the values of $\Delta\kappa_s$ are plotted as a function of PPG mass fraction at various salt concentrations at 298.15 K. Figure 6 shows that, at each temperature at a constant polymer concentration, the values of $\Delta\kappa_s$ become less negative as the concentration of salt increases.

Conclusions

Experimental data at $T = (288.15 \text{ to } 313.15)$ K of density and ultrasonic velocity of PPG in water + tri-potassium citrate have been reported. The values of excess specific volumes and isentropic compressibilities of solutions were calculated from the measured data. The excess specific volumes are negative and decrease in magnitude as temperature or concentration of tri-potassium citrate increases. The values of κ_s show a shallow

minimum that becomes more distinct and shows a tendency to shift to a higher PPG concentration as the temperature or concentration of tri-potassium citrate is lowered. Also it was found that, for the binary aqueous K_3Cit solutions and ternary solutions with low concentration of PPG, the measured values of κ_s decrease with the increase in temperature, which is completely opposite that for high PPG concentrations and pure PPG. In general it has been found that the effect of concentration of tri-potassium citrate on the volumetric behavior of aqueous PPG solutions is similar to the effect of temperature on the volumetric behavior of aqueous PPG solutions.

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