Application of Flory Theory to Mixtures of Molten Tetrahydrates of Calcium and Cadmium Nitrates

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The Flory theory has successfully been employed to predict the excess volume, viscosity of mixtures, surface tension, and the ultrasonic velocities of mixtures of molten tetrahydrates of calcium and cadmium nitrates. The agreement in the computed and the experimental values has been good. The excess volume as well as the excess adiabatic compressibility suggest maximum solute-solvent interactions at ca. 0.55 mole fraction of Cd^{2+}.

Even though the Flory theory[1,2] was meant to explain the behavior of nonpolar liquids and their mixtures, it was adequately employed to evaluate[1-5] the reduced and the characteristic thermodynamic parameters of polar liquids as well. These parameters, in turn, were employed to evaluate the excess volume, the excess thermodynamic properties, the viscosity of mixtures, the excess viscosity of mixing, the surface tension,[4,5,9,11-13] and the ultrasonic velocity of low melting salts.

In view of the interdependence/correlation of these properties with the intermolecular interactions, they were employed to highlight the solute-solvent interactions. For this purpose, molten tetrahydrates of calcium and cadmium nitrates as well as their mixtures were chosen.

Experimental

Commercial tetrahydrates of calcium and cadmium nitrates (BDH, E. Mercks) were recrystallized. The water of crystallization was determined by the method of comparison of their measured densities with those of the reported values.[14,15]

The density and viscosity measurements were made using a pycnometer (±0.03%) and a Cannon-Ubbelohde viscometer[16] (± 0.3%) in an ultrathermostat (type u-10) of ±0.01 °C thermal stability. The surface tension was measured with a stalagmometer for the purpose of comparison with the predicted values. The ultrasonic velocity (within ± 0.15%) was measured using an ultrasonic interferometer (NPL patent, Mittal's M 77).

Results and Discussion

The Excess Volume. The reduced and the characteristic thermodynamic variables ($\tilde{V}$, $\tilde{T}$ and V^*, T^*, P^*) of Flory theory[1-3] obtained essentially from the density data are employed for the evaluation of excess/reduced volume, $\tilde{V}^E$ using the relevant relations given elsewhere. The computed values of the excess

Table 1. Experimental and Computed Values of Excess Volume (V^E/ml·mol^{-1}) of Molten $Ca(NO_3)_2 \cdot 4.31\ H_2O$-$Cd(NO_3)_2 \cdot 4.34\ H_2O$ System

Mole fraction of Cd^{2+}		T/K=303.15	308.15	313.15	318.15	323.15	328.15	333.15
0.063	1[a]	−0.1258	−0.1408	−0.1550	−0.1692	−0.1834	−0.1976	−0.2118
	2	−0.1261	−0.1414	−0.1563	−0.1702	−0.1843	−0.1996	−0.2142
0.119	1	−0.2506	−0.2736	−0.2966	−0.3196	−0.3426	−0.3656	−0.3886
	2	−0.2519	−0.2708	−0.2915	−0.3161	−0.3403	−0.3658	−0.3910
0.168	1	−0.3548	−0.3864	−0.4181	−0.4497	−0.4814	−0.5130	−0.5447
	2	−0.3550	−0.3829	−0.4116	−0.4450	−0.4781	−0.5118	−0.5462
0.221	1	−0.4688	−0.5062	−0.5434	−0.5806	−0.6178	−0.6550	−0.6922
	2	−0.4706	−0.5037	−0.5385	−0.5769	−0.6149	−0.6551	−0.6948
0.300	1	−0.6877	−0.7165	−0.7453	−0.7741	−0.8029	−0.8317	−0.8605
	2	−0.6875	−0.7118	−0.7378	−0.7682	−0.7970	−0.8288	−0.8610
0.323	1	−0.7395	−0.7676	−0.7957	−0.8238	−0.8519	−0.8800	−0.9081
	2	−0.7394	−0.7636	−0.7886	−0.8178	−0.8465	−0.8783	−0.9085
0.362	1	−0.8397	−0.8661	−0.8924	−0.9188	−0.9451	−0.9715	−0.9979
	2	−0.8413	−0.8631	−0.8866	−0.9143	−0.9404	−0.9700	−0.9989
0.432	1	−1.0090	−1.0249	−1.0408	−1.0567	−1.0726	−1.0885	−1.1044
	2	−1.0100	−1.0217	−1.0350	−1.0514	−1.0670	−1.0869	−1.1058
0.554	1	−1.1801	−1.1932	−1.2063	−1.2194	−1.2325	−1.2455	−1.2587
	2	−1.1813	−1.1910	−1.2011	−1.2142	−1.2261	−1.2436	−1.2610
0.599	1	−1.1881	−1.2023	−1.2164	−1.2306	−1.2447	−1.2589	−1.2730
	2	−1.1899	−1.1994	−1.2114	−1.2251	−1.2386	−1.2573	−1.2746
0.677	1	−1.1497	−1.1684	−1.1870	−1.2057	−1.2243	−1.2430	−1.2616
	2	−1.1496	−1.1634	−1.1794	−1.1982	−1.2146	−1.2387	−1.2615
0.739	1	−1.1003	−1.1134	−1.1265	−1.1396	−1.1527	−1.1658	−1.1789
	2	−1.1004	−1.1097	−1.1192	−1.1323	−1.1439	−1.1619	−1.1794

a) 1. Experimental values of V^E. 2. Computed values of V^E.

Table 2. Comparison of Experimental and Computed Viscosity ($\eta \times 10$/N s m^{-2}) of Molten $Ca(NO_3)_2 \cdot 4.31\ H_2O$-$Cd(NO_3)_2 \cdot 4.34\ H_2O$ System

Mole fraction of Cd^{2+}	T/K= 303.15	308.15	313.15	318.15	323.15	328.15	333.15
0.000	1[a)]	1.389	1.064	0.827	0.649	0.521	0.427	0.360
0.063	1	1.370	1.043	0.812	0.640	0.520	0.420	0.334
	2	1.322	0.997	0.799	0.632	0.510	0.422	0.356
0.119	1	1.291	1.003	0.790	0.630	0.509	0.440	0.363
	2	1.255	0.978	0.773	0.616	0.499	0.416	0.352
0.168	1	1.249	0.971	0.771	0.617	0.500	0.421	0.360
	2	1.214	0.952	0.758	0.607	0.493	0.414	0.351
0.221	1	1.201	0.949	0.751	0.611	0.501	0.421	0.356
	2	1.180	0.931	0.745	0.600	0.487	0.411	0.348
0.300	1	1.181	0.941	0.751	0.601	0.491	0.420	0.356
	2	1.170	0.922	0.736	0.590	0.478	0.403	0.339
0.323	1	1.171	0.938	0.749	0.600	0.485	0.417	0.351
	2	1.168	0.920	0.735	0.588	0.477	0.402	0.338
0.362	1	1.167	0.921	0.737	0.591	0.475	0.400	0.332
	2	1.166	0.918	0.732	0.585	0.473	0.398	0.332
0.432	1	1.165	0.917	0.733	0.589	0.482	0.408	0.340
	2	1.164	0.916	0.729	0.581	0.469	0.394	0.329
0.554	1	1.162	0.926	0.732	0.591	0.470	0.400	0.330
	2	1.126	0.888	0.708	0.565	0.456	0.383	0.320
0.599	1	1.131	0.890	0.714	0.563	0.461	0.390	0.320
	2	1.092	0.863	0.689	0.551	0.446	0.375	0.313
0.677	1	1.070	0.853	0.677	0.561	0.452	0.380	0.314
	2	1.019	0.810	0.650	0.522	0.424	0.358	0.300
0.739	1	1.021	0.830	0.661	0.534	0.440	0.367	0.311
	2	0.967	0.769	0.616	0.495	0.403	0.340	0.285
1.000	1	0.588	0.476	0.385	0.315	0.261	0.220	0.188

a) 1. Experimental values of η. 2. Computed values of η.

volume agree well with those obtained experimentally ($V^E = V - \sum_i x_i V_i$) (Table 1). The minimum value of V^E corresponds to 0.55—0.599 mole fraction of Cd^{2+} at every temperature of measurement within the range, 303.15 to 333.15 K.

The Viscosity of Mixture. The viscosities of binary mixtures have been obtained using the equation,[7-9]

$$\ln \eta_{min} = -\frac{\Delta G_m}{RT} + \frac{1}{\tilde{V}-1} - \sum_i \frac{x_i}{\tilde{V}_i - 1} + \sum_i x_i \ln \eta_i ,$$

in which

$$\Delta G_m = \sum_i x_i P_i^* V_i^* \left[\left(\frac{1}{\tilde{V}_i} + \frac{1}{\tilde{V}} \right) + 3\tilde{T}_i \ln \left(\frac{\tilde{V}_i^{1/3} - 1}{\tilde{V}^{1/3} - 1} \right) \right] + x_1 V_1^* \theta_2 x_{12} V .$$

An examination of Table 2 reveals a reasonably good agreement between the experimental and the computed values of viscosity. The viscosity has been found to decrease with an increase in the mole fraction of Cd^{2+} as well as with temperature.

The Excess Adiabatic Compressibility. The excess adiabatic compressibility $\beta_S^E (= \beta_S - \sum_i x_i \beta_{Si})$ of molten Ca^{2+}-Cd^{2+} mixtures has been found to be negative, which suggests the presence of strong associative interactions between the solvent and the solute molecules. However, β_S^E is not affected significantly with temperature. The value of β_S^E shows variations with composition, as apparent from Fig. 1 in which the presence of minima at 0.554 mole fraction of Cd^{2+} indicates maximum interaction at this composition.

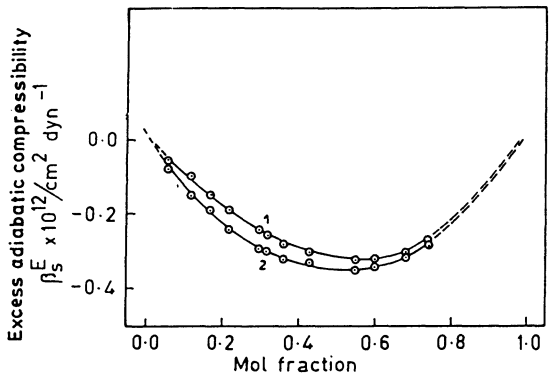

Fig. 1. Plot of excess adiabatic compressibility vs. [solute] for $Ca(NO_3)_2 \cdot 4.31 H_2O$-$Cd(NO_3)_2 \cdot 4.34 H_2O$ system at 1; 303.15 K and 2; 333.15 K.

It is noteworthy that the excess volume of mixing and the excess adiabatic compressibility factor, both of them suggest the maximum solvent-solute interaction at ca. 0.55 mole fraction of Cd^{2+}.

The Surface Tension. The agreement between the predicted values of surface tension, σ obtainable essentially from the Flory theory[4,5,10,12] with those obtained experimentally has been reasonably good even though σ was measured using the stalagmometer. An examination of Table 3 reveals that unlike the pure $Cd(NO_3)_2 \cdot 4H_2O$ Auerbach relation does not seem to apply to Ca^{2+}-Cd^{2+} system in which Ca^{2+} may appar-

Table 3. Experimental and Computed Values of Surface Tension (σ/dyn cm^{-1}) of Molten $Ca(NO_3)_2 \cdot 4.31\ H_2O$-$Cd(NO_3)_2 \cdot 4.34\ H_2O$ System

Mole fraction of Cd^{2+}		T/K=303.15	308.15	313.15	318.15	323.15	328.15	333.15
0.000	1[a)]	95.41	94.68	94.01	93.28	92.71	91.87	91.21
	2	94.42	93.98	93.70	93.01	92.68	91.29	91.27
	3	74.48	77.82	77.02	76.30	75.51	74.83	74.12
0.063	1	95.66	94.87	94.22	93.50	92.82	92.10	91.35
	2	94.64	94.19	93.89	93.21	92.92	91.57	91.55
	3	79.85	79.12	78.33	77.61	76.87	76.09	75.38
0.119	1	95.88	95.17	94.36	93.65	92.90	92.20	91.50
	2	94.77	94.29	94.01	93.34	93.08	92.56	91.72
	3	81.04	80.36	79.57	78.82	78.07	77.27	76.55
0.168	1	96.15	95.34	94.64	93.85	93.13	92.40	91.62
	2	94.71	94.27	93.94	93.27	93.04	92.51	91.70
	3	82.20	81.45	80.65	79.88	79.09	78.30	77.56
0.221	1	96.51	95.70	94.84	94.16	93.28	92.53	91.72
	2	94.74	94.28	93.95	93.26	93.05	91.79	91.71
	3	83.38	82.60	81.81	81.00	80.20	79.41	78.64
0.300	1	96.90	96.00	95.25	94.40	93.62	92.41	92.01
	2	95.40	94.89	94.46	93.75	93.10	92.88	92.09
	3	85.19	84.26	83.54	82.72	81.87	81.08	80.24
0.323	1	97.14	96.30	95.40	94.56	93.73	92.90	92.14
	2	95.15	94.99	94.55	93.83	93.15	92.95	92.15
	3	85.71	84.76	84.03	83.20	82.35	81.56	80.7
0.362	1	97.36	96.50	95.58	94.83	93.98	93.16	92.24
	2	95.71	95.16	94.69	93.95	93.72	93.03	92.24
	3	86.61	85.76	84.88	84.03	83.16	82.35	81.48
0.432	1	97.87	97.01	96.00	95.20	94.20	93.41	92.50
	2	96.39	95.79	95.24	94.45	94.20	93.44	92.64
	3	88.21	87.34	86.43	85.53	84.65	83.77	82.88
0.554	1	98.86	97.82	96.97	95.90	95.01	94.04	93.03
	2	97.13	96.45	95.79	94.95	94.72	93.83	93.06
	3	91.02	90.05	89.09	88.58	87.18	86.24	85.30
0.599	1	99.27	98.30	97.20	96.30	95.21	94.32	93.30
	2	97.52	96.84	96.17	95.33	95.10	93.85	93.44
	3	92.03	91.04	90.05	89.08	88.11	87.15	86.19
0.677	1	100.03	98.90	97.95	96.76	95.83	94.70	93.70
	2	97.84	97.14	96.43	95.59	95.37	94.44	93.70
	3	93.78	92.77	91.74	90.71	89.69	88.67	86.69
0.739	1	100.60	99.55	98.37	97.35	96.20	95.20	94.80
	2	99.06	98.32	97.56	96.68	96.47	95.47	94.73
	3	95.09	94.02	92.97	91.91	91.03	89.83	88.79
1.000	1	103.50	102.16	100.80	99.55	98.60	96.90	95.50
	2	100.26	99.42	98.49	97.54	97.36	96.17	95.47
	3	100.10	98.95	97.79	96.63	95.47	94.32	93.18

a) 1. Experimental values of σ. 2. Computed values of σ using Flory theory. 3. Computed values of σ using Auerbach relation.

Table 4. Experimental and Computed Ultrasonic Velocity (U/m$\cdot$s^{-1}) in Molten $Ca(NO_3)_2 \cdot 4.31\ H_2O$-$Cd(NO_3)_2 \cdot 4.34\ H_2O$ System

Mole fraction of Cd^{2+}		T/K=303.15	308.15	313.15	318.15	323.15	328.15	333.15
0.000	1[a)]	1738.11	1730.11	1721.61	1713.50	1704.21	1696.71	1688.51
	5	1965.99	1962.85	1961.99	1955.33	1953.75	1937.12	1939.89
0.063	1	1736.60	1728.20	1719.81	1711.40	1703.01	1694.60	1686.21
	2	1738.06	1730.54	1721.25	1721.98	1703.58	1695.87	1687.51
	3	1734.44	1726.99	1717.74	1709.52	1700.17	1692.52	1684.21
	4	1739.62	1732.22	1723.30	1714.86	1705.72	1698.19	1690.03
	5	1951.00	1947.94	1946.94	1940.66	1939.70	1923.99	1926.84
0.119	1	1735.11	1727.32	1718.83	1710.35	1701.87	1693.38	1684.91
	2	1738.03	1730.31	1720.31	1712.51	1703.03	1695.13	1686.62
	3	1731.62	1724.02	1714.72	1706.39	1696.99	1689.21	1680.79
	4	1741.12	1733.62	1724.69	1716.22	1707.08	1699.46	1691.25
	5	1931.94	1928.60	1927.98	1921.94	1921.58	1917.62	1909.13
0.168	1	1735.01	1726.51	1717.91	1709.31	1700.71	1692.11	1683.51
	2	1738.01	1730.11	1720.66	1712.11	1702.55	1694.48	1685.84

Table 4. (Continued)

Mole fraction of Cd^{2+}		T/K=303.15	308.15	313.15	318.15	323.15	328.15	333.15
	3	1729.43	1721.71	1712.36	1703.94	1694.48	1686.58	1678.07
	4	1742.38	1734.81	1725.89	1717.41	1708.27	1700.58	1692.33
	5	1912.95	1910.14	1908.88	1902.91	1902.99	1898.88	1890.98
0.221	1	1734.50	1725.82	1717.13	1708.45	1699.77	1691.08	1682.40
	2	1737.97	1729.89	1720.37	1711.66	1702.03	1693.81	1685.02
	3	1727.46	1719.59	1710.19	1701.65	1692.15	1684.10	1675.49
	4	1743.75	1736.05	1727.10	1718.54	1709.38	1701.58	1693.24
	5	1894.35	1891.40	1890.10	1884.02	1884.32	1870.44	1872.48
0.300	1	1733.61	1724.75	1715.91	1707.05	1698.21	1689.35	1680.50
	2	1737.97	1729.56	1719.92	1711.00	1701.25	1692.75	1683.77
	3	1727.46	1717.01	1707.52	1698.81	1689.21	1680.95	1672.17
	4	1743.75	1738.31	1729.09	1720.22	1710.82	1702.63	1692.98
	5	1871.47	1867.92	1865.48	1859.25	1853.84	1854.13	1847.27
0.323	1	1733.40	1724.51	1715.60	1706.71	1697.81	1688.91	1680.01
	2	1737.91	1729.47	1719.81	1710.81	1701.03	1692.45	1683.41
	3	1724.57	1716.41	1706.89	1698.13	1688.50	1680.18	1671.35
	4	1747.03	1738.84	1729.57	1720.63	1711.19	1702.92	1694.18
	5	1861.70	1862.65	1859.94	1853.59	1847.66	1848.06	1840.48
0.362	1	1733.31	1724.28	1715.27	1706.25	1697.23	1688.22	1679.21
	2	1737.88	1729.31	1719.58	1710.49	1700.65	1691.95	1682.81
	3	1723.81	1715.53	1705.97	1697.12	1687.45	1679.01	1670.11
	4	1748.24	1739.88	1730.52	1721.46	1711.92	1703.49	1694.62
	5	1854.88	1850.76	1847.65	1841.06	1841.04	1834.98	1827.63
0.432	1	1733.00	1723.84	1714.67	1705.51	1696.34	1687.18	1678.01
	2	1737.83	1729.03	1719.20	1709.92	1699.97	1691.04	1681.72
	3	1722.91	1714.41	1704.75	1695.73	1685.97	1677.33	1668.25
	4	1750.27	1741.53	1731.91	1722.54	1712.75	1703.95	1694.78
	5	1840.95	1836.27	1832.27	1825.09	1824.85	1817.99	1810.63
0.554	1	1733.10	1723.51	1714.01	1704.51	1695.50	1685.50	1676.00
	2	1737.76	1728.53	1718.32	1708.92	1698.86	1689.46	1679.84
	3	1722.72	1713.81	1703.99	1694.66	1684.72	1675.69	1666.31
	4	1752.29	1743.07	1733.19	1723.48	1713.45	1704.20	1694.67
	5	1809.72	1804.21	1798.91	1790.32	1791.37	1783.07	1776.18
0.599	1	1733.10	1723.51	1713.90	1704.31	1694.70	1685.11	1675.50
	2	1737.74	1728.35	1718.29	1708.55	1698.36	1688.88	1679.14
	3	1723.11	1714.04	1704.15	1694.69	1684.68	1675.50	1666.01
	4	1752.35	1742.98	1733.23	1723.23	1713.14	1703.76	1694.12
	5	1801.40	1795.97	1790.57	1783.08	1783.11	1770.40	1768.19
0.677	1	1733.71	1723.85	1714.00	1704.15	1694.30	1684.45	1674.60
	2	1737.69	1728.03	1717.85	1707.91	1697.61	1687.87	1677.93
	3	1724.37	1715.00	1704.99	1695.31	1685.17	1675.71	1665.99
	4	1751.80	1742.22	1732.20	1722.26	1712.10	1702.52	1692.73
	5	1779.07	1773.55	1767.86	1760.49	1760.76	1752.26	1746.06
0.739	1	1734.30	1724.28	1714.27	1704.25	1696.23	1684.22	1674.20
	2	1737.66	1727.78	1717.51	1707.41	1697.02	1687.08	1676.98
	3	1725.88	1716.28	1706.16	1696.28	1686.04	1676.35	1666.45
	4	1751.14	1741.27	1731.08	1720.94	1710.62	1700.78	1690.77
	5	1782.25	1776.43	1770.33	1762.71	1763.22	1754.07	1748.04
1.000	1	1737.50	1726.74	1716.10	1705.31	1694.54	1683.77	1673.01
	5	1739.11	1732.33	1724.40	1716.23	1717.07	1705.93	1700.57

a) 1. Experimental U. 2. Computed U using.[18,21] 3. Computed U using.[19,22] 4. Computed U using.[20,23] 5. Computed U using Flory theory.

ently be responsible for offsetting the agreement between the computed and the experimental values. Also, the agreement between the two values is much better at higher temperatures compared to those at lower ones. The σ values of $Ca(NO_3)_2 \cdot 4.3H_2O$ and $Ca(NO_3)_2 \cdot 4.1H_2O$ are, respectively, 1.1% and 0.32% lower when compared with that of $Ca(NO_3)_2 \cdot 4H_2O$.[17] Such a difference seems to have been caused by the difference in the water of hydration which seems to be responsible for decreasing the corresponding cohesive forces.

The Ultrasonic Velocity. The computed values of ultrasonic velocity based on the relation,[11]

$$U = \left\{ \left[(kT^*P^{*2})^{1/3} \left(M\tilde{V}^{-5/3} - \frac{\tilde{V}^{1/3}-1}{\tilde{V}^2} \ln \frac{V^{1/3}-0.5}{V^{1/3}-1} \right) \right] / \rho \times 6.3 \times 10^{-4} \right\}^{2/3}$$

on comparison with the experimental values reveal that the agreement between the two values is better in the case of pure component of Cd^{2+}. Nevertheless, an excellent agreement between the computed values obtained using essentially Nomoto,[18] Van Dael et al.,[19] and Schaaff's collision factor theory[20] and the experimental values of ultrasonic velocities may be seen from Table 4.

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