Excess Volumes of Binary Mixtures That Contain Olive Oil with Alkyl and Vinyl Acetates

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ABSTRACT: This paper reports densities and excess molar volumes for ethyl acetate, vinyl acetate, propyl acetate, isopropyl acetate, and butyl acetate with olive oil at temperatures from 283.15–298.15 K. Redlich-Kister polynomials were fitted to the results of excess volumes. All the systems showed slight deviations from ideality. The excess volumes decreased with the number of carbon atoms of the acetate, but increased with an increase of temperature.

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KEY WORDS: Alkyl acetate, excess molar volumes, olive oil, vinyl acetate.

In this paper, we investigated the interaction between alkyl and vinyl acetates with olive oil by measuring the excess molar volumes of mixing. In previous works, we presented experimental densities and excess molar volumes of mixtures of alkanes with sunflower seed oil (1) and alkanes and alcohols with olive oil (2). There are other reports on the thermodynamic properties of these kinds of mixtures (3–5), although the temperatures, oils, and solvents were different.

This is a continuation of our research on the physical properties of organic solvents–vegetable oil systems. Natural oils are composed of mixtures of triglycerides containing mainly oleic and linoleic acids. The mixture of the fatty oils with organic solvents changes the physical properties of the mixture, depending on the structure of the solvent. Some properties are interesting in the refining-oil plant design because oils are mixed with solvents like hexane or acetone in several stages. In the new technologies for extraction and winterizing, some other solvents are being studied. In this paper, we present densities and excess molar volumes of mixtures formed by ethyl acetate, vinyl acetate, propyl acetate, isopropyl acetate, and butyl acetate with olive oil at temperatures from 283.15–298.15 K.

EXPERIMENTAL PROCEDURES

Analytical grade ethyl acetate, vinyl acetate, propyl acetate, isopropyl acetate, and butyl acetate were obtained from Fluka (Buchs, Switzerland) with a purity of >99%. Refined olive oil was supplied by Koipe (Jaén, Spain) and it was previously analyzed to determine its composition. The contents of water and of volatile compounds were less than 0.1%. The fatty acid composition was analyzed by means of a PerkinElmer Sigma 3B gas chromatograph equipped with a flame detector (Norwalk, CT). The chromatographic technique and the chemical procedure for the preparation of fatty acids were described previously (6). The oil composition obtained was palmitic acid, 12.4%; stearic acid, 2.8%; oleic acid, 77.8%; and linoleic acid, 7%. The uncertainty in mole percentage for these results was better than 0.1%. From this composition, the average molar mass of this oil has been computed in accordance with the following expression:

$$M_{\text{oil}} = 3\left(\sum_{i-1}^{N} x_i M_i\right) + M_{CH-C-CH} \quad [1]$$

where x_i is the molar fraction and M_i is the molar mass of each fatty acid contributing to the concentration analysis. N is the number of fatty acids found by analysis, and $M_{\text{CH-C-CH}}$ is the molar mass contribution of the glycerol molecule fraction. The computed average molar mass in olive oil samples was 875.84 g mol^{-1}. The variation in the composition, due to different samples, affects mainly the mono- and polyunsaturated fatty acids. The change in molar mass is less than ±1 g mol^{-1} and in excess molar volume is lower than $\pm 2 \times 10^{-3}$ $\text{cm}^3\ \text{mol}^{-1}$.

Mixtures of the required composition were prepared by mass using a Salter ER-182A balance, taking precautions to prevent evaporation. The accuracy is within $\pm 5 \times 10^{-4}$ g. The possible error in the mole fraction is estimated to be less than ±0.0002. The densities (ρ) were measured using an Anton Paar DMA-58 (Graz, Austria) vibrating-tube densitometer with a resolution of $\pm 1 \times 10^{-5}$ g·cm^{-3}. The densitometer was calibrated with water and air, using the corresponding density of water at each temperature (7), and air density was calculated by Equation 2:

$$\rho_{t,P} = \frac{0.0012930 \cdot P}{1 + 0.00367 \cdot t} \quad [2]$$

where t is temperature in °C and P is pressure in atm. The glass oscillator was inserted into a glass tube and a copper housing. A semiconductor Peltier element, which is connected to the copper housing, ensured sufficient thermostating of both the sample and oscillator tube. A resistance ther-

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TABLE 1
Refractive Indices and Densities (g·cm^{-3}) of the Used Solvents and Oil[a]

	Refractive index (298.15 K)		Density (298.15)		Density (293.15 K)		Density (288.15 K)		Density (283.15 K)	
Compound	Lit.	Exptl.	Lit.	Exptl.	Lit.	Exptl.	Lit.	Exptl.	Lit.	Exptl.
Ethyl acetate	1.36978	1.3697	0.89455	0.89430	0.90063	0.90038	—	0.90645	—	0.91243
Vinyl acetate	1.3934	1.3930	—	0.92565	0.9312	0.93204	—	0.93845	—	0.94475
Propyl acetate	1.3828	1.3824	0.88303	0.88206	—	0.88762	—	0.89315	—	0.89867
Isopropyl acetate	1.375	1.3753	0.8702	0.86645	0.8773	0.87227	—	0.87804	—	0.88379
Butyl acetate	1.3918	1.3920	0.87636	0.87605	—	0.88116	—	0.8863	—	0.89141
Olive oil	—	1.4672	—	0.90888	—	0.91228	—	0.91575	—	0.91924

[a]Lit., literature value (7); exptl., experimental value.

mometer measured the temperature of the cell and controlled the current of the Peltier element accordingly. Temperatures were accurate to $\pm 1 \times 10^{-2}$ K.

Excess volumes were accurate to $\pm 2 \times 10^{-3}$ cm^{-3} · mol^{-1}. Experimental densities and refractive indices of the used solvents at different temperatures, with the corresponding literature values (7), are listed in Table 1.

RESULTS AND DISCUSSION

Densities and excess molar volumes of the prepared mixtures are listed in Tables 2–6. The experimental densities ρ were converted into excess molar volumes V^E using the following equation:

$$V^E = (x_1 M_1 + x_2 M_2)/\rho - [(x_1 M_1/\rho_1) + (x_2 M_2/\rho_2)] \quad [3]$$

where x_1 is the mole fraction of the more volatile compound, x_2 is the mole fraction of the olive oil, M_1 and M_2 are the molar masses of the solvent and oil, respectively, and ρ_1 and ρ_2 are the densities of the pure components. The excess volume data were correlated as a function of composition using a three constant Redlich-Kister expansion:

$$V^E,\ \text{in units of cm}^3\cdot\text{mol}^{-1} = x_1 x_2 \sum_{k\geq 0} a_k (x_2 - x_1)^k \quad [4]$$

where a_k are adjustable parameters obtained by the least squares method. Table 7 summarizes the values of the parameters a_k together with the standard deviations (σ) in V^E of the differences between values from Equation 4 and Equation 3. The parameters of Equation 4 were used to calculate the solid curves in Figures 1 and 2. In Figure 1, V^E of the acetates + olive oil mixtures are plotted vs. the mole fraction of acetate. Figure 2 shows V^E of isopropyl acetate + olive oil mixtures vs. the mole fraction of isopropyl acetate at different temperatures.

V^E is a thermodynamic function that measures the mixture's deviation from ideal behavior. In accordance with the

TABLE 2
Experimental Densities ρ (g·cm^{-3}) and Excess Volumes V^E (cm^4·mol^{-1}) for Ethyl Acetate + Olive Oil Mixtures at Several Temperatures

	T = 298.15 K		T = 293.15 K		T = 288.15 K		T = 283.15 K	
x_1[a]	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0.0445	0.90878	0.032	0.91219	0.034	0.91566	0.046	0.91914	0.068
0.0989	0.90866	0.056	0.91208	0.066	0.91557	0.074	0.91906	0.099
0.1624	0.90851	0.078	0.91195	0.089	0.91544	0.116	0.91894	0.149
0.2141	0.90838	0.090	0.91184	0.105	0.91533	0.143	0.91883	0.187
0.2649	0.90823	0.107	0.91170	0.127	0.91519	0.181	0.91873	0.213
0.3122	0.90806	0.132	0.91156	0.147	0.91507	0.203	0.91861	0.247
0.3576	0.90789	0.148	0.91141	0.165	0.91494	0.221	0.91849	0.272
0.4036	0.90769	0.167	0.91124	0.183	0.91479	0.241	0.91835	0.297
0.4484	0.90747	0.185	0.91106	0.195	0.91464	0.250	0.91822	0.311
0.4967	0.90721	0.197	0.91083	0.212	0.91445	0.262	0.91805	0.328
0.5528	0.90687	0.200	0.91054	0.217	0.91419	0.275	0.91783	0.340
0.5944	0.90658	0.198	0.91029	0.218	0.91396	0.285	0.91764	0.348
0.6560	0.90605	0.197	0.90985	0.214	0.91358	0.284	0.91732	0.348
0.7044	0.90554	0.189	0.90941	0.211	0.91321	0.279	0.91702	0.341
0.7549	0.90486	0.182	0.90883	0.207	0.91271	0.278	0.91664	0.328
0.7962	0.90415	0.173	0.90823	0.199	0.91223	0.262	0.91627	0.307
0.8511	0.90287	0.159	0.90715	0.187	0.91137	0.238	0.91559	0.282
0.9001	0.90120	0.141	0.90577	0.165	0.91029	0.204	0.91478	0.241
0.9500	0.89858	0.107	0.90367	0.119	0.90867	0.147	0.91366	0.167

[a]Mole fraction of ethyl acetate.

TABLE 3
Experimental Densities ρ (g·cm^{-3}) and Excess Volumes V^E (cm^4·mol^{-1}) for Vinyl Acetate + Olive Oil Mixtures at Several Temperatures

	T = 298.15 K		*T* = 293.15 K		*T* = 288.15 K		*T* = 283.15 K	
x_1[a]	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0.0444	0.90892	0.035	0.91233	0.038	0.91580	0.051	0.91928	0.072
0.1032	0.90901	0.052	0.91243	0.063	0.91591	0.083	0.91938	0.129
0.1443	0.90906	0.078	0.91249	0.097	0.91598	0.119	0.91947	0.158
0.2022	0.90916	0.104	0.91259	0.138	0.91609	0.169	0.91960	0.206
0.2523	0.90925	0.130	0.91269	0.172	0.91621	0.203	0.91974	0.238
0.3085	0.90937	0.155	0.91283	0.200	0.91636	0.242	0.91991	0.279
0.3672	0.90952	0.177	0.91300	0.228	0.91656	0.270	0.92012	0.316
0.4129	0.90966	0.190	0.91316	0.244	0.91674	0.294	0.92033	0.339
0.4527	0.90980	0.200	0.91333	0.255	0.91693	0.301	0.92053	0.354
0.5094	0.91004	0.213	0.91361	0.266	0.91725	0.313	0.92087	0.376
0.5488	0.91022	0.225	0.91382	0.281	0.91751	0.321	0.92116	0.385
0.6013	0.91052	0.237	0.91418	0.289	0.91793	0.327	0.92161	0.399
0.6434	0.91081	0.247	0.91453	0.296	0.91835	0.329	0.92207	0.400
0.6986	0.91131	0.247	0.91511	0.299	0.91901	0.333	0.92281	0.403
0.7510	0.91193	0.250	0.91582	0.305	0.91983	0.337	0.92376	0.394
0.8006	0.91273	0.244	0.91675	0.300	0.92092	0.328	0.92496	0.386
0.8522	0.91392	0.232	0.91816	0.281	0.92254	0.308	0.92682	0.351
0.9010	0.91563	0.213	0.92022	0.249	0.92490	0.274	0.92952	0.302
0.9567	0.91930	0.142	0.92453	0.167	0.92987	0.181	0.93512	0.198

[a]Mole fraction of vinyl acetate.

already accepted hypothesis for the liquid estate theories, it could be interpreted as a manifestation of the variations of the intermolecular forces of the packing of all the component molecules. These variations will depend on the molecular structure: polar groups, size, and geometry.

The ester group is relatively polar. Owing to the inductive effect due to the oxygen, the methyl group of the acetate will also have a small, partial positive charge, as well as the alkyl chain joined to the noncarboxylic oxygen. Olive oil molecules are triglycerides (more than 99%). The difference with the simple esters is that they present two slightly different zones: a central polar axis built up by three ester groups (alkoxycarboxylic) and three long nonpolar chains. The molecule is voluminous, but it is not compact, which means that there are hollows between the nonpolar chains where small molecules with low interaction could be introduced.

TABLE 4
Experimental Densities ρ (g·cm^{-3}) and Excess Volumes V^E (cm^4·mol^{-1}) for Propyl Acetate + Olive Oil Mixtures at Several Temperatures

	T = 298.15 K		*T* = 293.15 K		*T* = 288.15 K		*T* = 283.15 K	
x_1[a]	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0.0558	0.90869	0.001	0.91210	0.007	0.91557	0.021	0.91903	0.065
0.1005	0.90852	0.005	0.91194	0.014	0.91541	0.040	0.91888	0.085
0.1537	0.90829	0.016	0.91172	0.032	0.9152	0.063	0.91867	0.120
0.2038	0.90804	0.035	0.91149	0.049	0.91498	0.085	0.91845	0.154
0.2512	0.90778	0.050	0.91125	0.063	0.91476	0.097	0.91823	0.177
0.2956	0.90752	0.057	0.911005	0.073	0.91453	0.108	0.918	0.199
0.3411	0.90722	0.066	0.91073	0.081	0.91426	0.126	0.91775	0.213
0.4097	0.90671	0.072	0.91026	0.085	0.91381	0.139	0.91733	0.224
0.4403	0.90645	0.074	0.91002	0.087	0.91358	0.145	0.91711	0.232
0.4917	0.90596	0.077	0.90957	0.089	0.91316	0.147	0.91671	0.237
0.5523	0.90528	0.076	0.90894	0.089	0.91257	0.148	0.91616	0.236
0.6045	0.90458	0.069	0.90829	0.085	0.91196	0.147	0.9156	0.228
0.6537	0.90379	0.059	0.90755	0.081	0.91128	0.137	0.91497	0.216
0.6989	0.90291	0.050	0.90673	0.075	0.91052	0.130	0.91428	0.200
0.7478	0.90173	0.038	0.90563	0.066	0.9095	0.120	0.91335	0.183
0.8004	0.90007	0.028	0.9041	0.055	0.90809	0.105	0.91205	0.165
0.8675	0.897	0.017	0.90126	0.044	0.90551	0.079	0.90967	0.135
0.9091	0.89419	0.011	0.89867	0.035	0.90313	0.066	0.90753	0.108
0.9546	0.88964	0.004	0.89449	0.023	0.89935	0.039	0.9041	0.073

[a]Mole fraction of propyl acetate.

TABLE 5
Experimental Densities ρ ($g·cm^{-3}$) and Excess Volumes V^E ($cm^4·mol^{-1}$) for Isopropyl Acetate + Olive Oil Mixtures at Several Temperatures

	T = 298.15 K		T = 293.15 K		T = 288.15 K		T = 283.15 K	
x_1[a]	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0.0525	0.90860	–0.005	0.91201	0.002	0.91549	0.008	0.91897	0.029
0.1033	0.90830	–0.012	0.91172	0.005	0.91521	0.018	0.91869	0.054
0.1521	0.90799	–0.019	0.91141	0.012	0.91492	0.027	0.91840	0.073
0.1942	0.90769	–0.022	0.91112	0.015	0.91463	0.037	0.91813	0.085
0.2477	0.90727	–0.027	0.91072	0.017	0.91424	0.045	0.91775	0.101
0.3053	0.90676	–0.034	0.91023	0.015	0.91377	0.054	0.91729	0.115
0.3611	0.90620	–0.045	0.90968	0.012	0.91324	0.061	0.91680	0.121
0.4066	0.90568	–0.054	0.90918	0.010	0.91275	0.067	0.91634	0.124
0.4486	0.90514	–0.062	0.90866	0.005	0.91225	0.066	0.91586	0.125
0.4947	0.90446	–0.071	0.90802	–0.005	0.91163	0.064	0.91527	0.124
0.5436	0.90363	–0.081	0.90723	–0.013	0.91087	0.060	0.91455	0.119
0.5940	0.90262	–0.094	0.90627	–0.025	0.90996	0.047	0.91367	0.113
0.6449	0.90141	–0.113	0.90512	–0.044	0.90885	0.035	0.91261	0.104
0.6989	0.89980	–0.125	0.90360	–0.060	0.90741	0.016	0.91124	0.086
0.7457	0.89804	–0.133	0.90193	–0.069	0.90582	0.007	0.90976	0.065
0.8010	0.89533	–0.144	0.89934	–0.075	0.90338	–0.008	0.90746	0.046
0.8515	0.89192	–0.144	0.89612	–0.082	0.90035	–0.024	0.90460	0.027
0.9066	0.88640	–0.122	0.89091	–0.070	0.89546	–0.027	0.90000	0.016
0.9520	0.87929	–0.081	0.88425	–0.048	0.88920	–0.018	0.89415	0.010

[a]Mole fraction of isopropyl acetate.

Most of the mixtures of olive oil with acetates showed positive V^E. They were only slightly negative with isopropyl acetate at 293.15 and 298.15 K, and with butyl acetate at 298.15 K. These results can be rationalized because pure ester molecules are closely packed due to dipole–dipole interactions. The packing is better for vinyl acetate, which has the highest density and the smallest molecular volume. When vinyl acetate is mixed with olive oil, the nonpolar chains partly break the connection among acetate molecules, increasing the intermolecular distances and causing positive V^E. Isopropyl acetate, on the other hand, has the lowest packing (and lowest density) of the acetates studied due to the methyl branch. When mixed with the olive oil, the isopropyl molecules may fit in the voids created by the nonpolar chains, producing a negative V^E.

In all the mixtures studied, V^E decreased when temperature increased. The molecular order stated by the dipole–dipole interactions was disturbed when the acetate molecules

TABLE 6
Experimental Densities ρ ($g·cm^{-3}$) and Excess Volumes V^E ($cm^3·mol^{-1}$) for Vinyl Acetate + Olive Oil Mixtures at Several Temperatures

	T = 298.15 K		T = 293.15 K		T = 288.15 K		T = 283.15 K	
x_1[a]	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0.0487	0.90865	–0.002	0.91205	0.011	0.91552	0.025	0.91901	0.036
0.0981	0.90840	–0.004	0.91180	0.021	0.91528	0.036	0.91877	0.057
0.1538	0.90809	–0.008	0.91149	0.029	0.91498	0.047	0.91848	0.076
0.2131	0.90771	–0.012	0.91112	0.038	0.91463	0.058	0.91814	0.093
0.2537	0.90743	–0.016	0.91084	0.041	0.91436	0.065	0.91788	0.100
0.3104	0.90699	–0.021	0.91042	0.042	0.91395	0.072	0.91749	0.108
0.3385	0.90675	–0.022	0.91018	0.040	0.91372	0.074	0.91727	0.109
0.4002	0.90616	–0.026	0.90962	0.033	0.91317	0.074	0.91675	0.113
0.4626	0.90545	–0.030	0.90895	0.026	0.91253	0.071	0.91612	0.117
0.5020	0.90493	–0.032	0.90846	0.021	0.91206	0.068	0.91567	0.116
0.5461	0.90428	–0.035	0.90784	0.016	0.91146	0.066	0.91510	0.115
0.5997	0.90334	–0.039	0.90695	0.010	0.91063	0.055	0.91429	0.109
0.6496	0.90230	–0.045	0.90595	0.007	0.90968	0.046	0.91339	0.101
0.7016	0.90098	–0.053	0.90468	0.002	0.90849	0.034	0.91225	0.090
0.7578	0.89918	–0.067	0.90295	–0.008	0.90685	0.023	0.91070	0.072
0.7948	0.89767	–0.069	0.90152	–0.015	0.90549	0.014	0.90942	0.060
0.8508	0.89468	–0.067	0.89870	–0.024	0.90283	0.001	0.90690	0.041
0.8994	0.89102	–0.061	0.89524	–0.027	0.89956	–0.006	0.90382	0.026
0.9535	0.88488	–0.046	0.88943	–0.021	0.89410	–0.012	0.89867	0.011

[a]Mole fraction of vinyl acetate.

TABLE 7
Adjustable Parameters in Equation 4 for Alkyl Acetate + Olive Oil at Four Different Temperatures

T (K)	a_0	a_1	a_2	a_3	σ^a ($cm^3 \cdot mol^{-1}$)
Ethyl acetate + oil					
298.15	0.7350	−0.3018	0.3610	−0.4701	0.0150
293.15	0.7955	−0.3129	0.5479	−0.6091	0.0107
288.15	1.0415	−0.3799	0.7115	−0.7951	0.0089
283.15	1.2837	−0.4891	0.8086	−0.6834	0.0120
Vinyl acetate + oil					
298.15	0.8327	−0.4050	0.8757	−1.0354	0.0103
293.15	1.0488	−0.3873	0.9990	−1.3852	0.0099
288.15	1.2195	−0.3153	1.0612	−1.5584	0.0116
283.15	1.4502	−0.5806	1.2569	−1.0494	0.0137
Propyl acetate + oil					
298.15	0.3080	0.0752	−0.3257	−0.1696	0.0019
293.15	0.3609	0.0418	−0.0927	−0.2867	0.0019
288.15	0.5864	−0.0645	0.0004	−0.2023	0.0026
283.15	0.9275	0.0327	0.2483	−0.3397	0.0065
Isopropyl acetate + oil					
298.15	−0.2781	0.4422	−0.7113	0.5499	0.0029
293.15	−0.0165	0.4011	−0.5300	0.2304	0.0020
288.15	0.2522	0.1814	−0.4741	0.1830	0.0017
283.15	0.4972	0.1435	−0.1910	0.1687	0.0024
Butyl acetate + oil					
298.15	−0.1206	0.1334	−0.3747	0.4410	0.0023
293.15	0.1034	0.2152	−0.1403	0.1763	0.0038
288.15	0.2776	0.1668	−0.1715	0.2130	0.0019
283.15	0.4651	0.0503	−0.0006	0.2900	0.0011

[a]Standard deviation of the differences between values calculated by Equation 4 and experimental data.

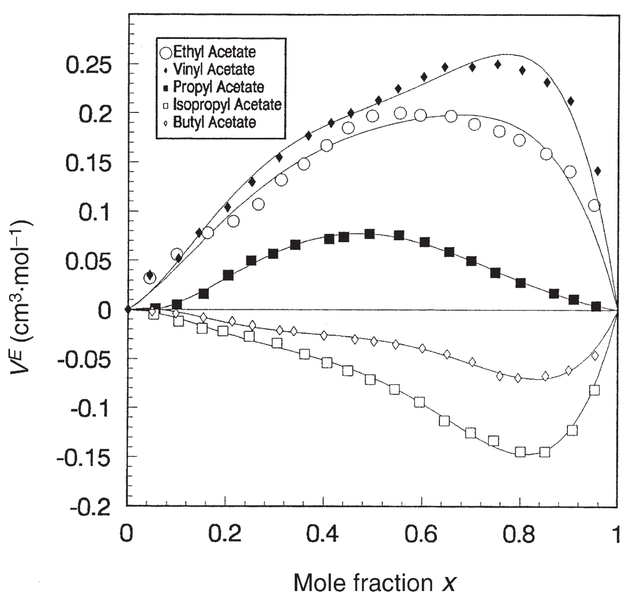

FIG. 1. Excess molar volumes (V^E) of mixtures of alkyl acetates with olive oil vs. mole fraction (x) of acetate. The solid lines correspond to values calculated by Equation 4.

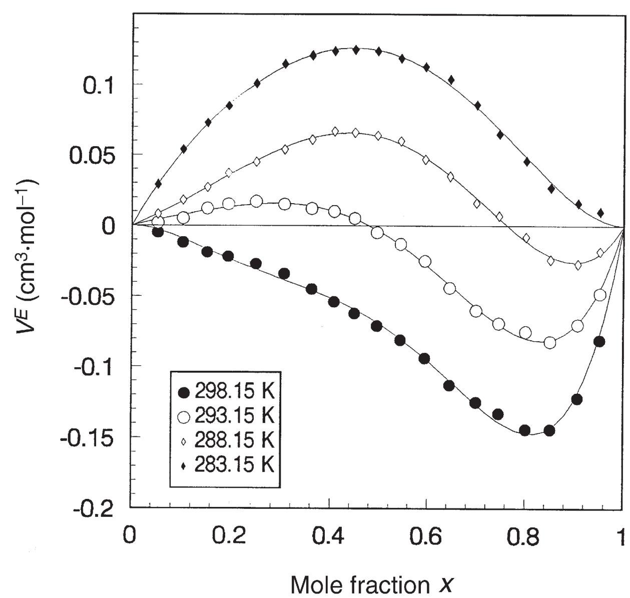

FIG. 2. Excess molar volumes of mixtures of isopropyl acetate with olive oil vs. mole fraction of acetate at different temperatures.

had more energy. Therefore, an addition of olive oil did not disturb as much as in a cooler, well-ordered structure. Therefore, V^E were smaller than the ones at lower temperatures.

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