Limiting Partial Molar Volumes of Tetra-*n*-alkylammonium Bromides in *x*Dimethylformamide + (1 − *x*)Water Mixtures at 298.15 K

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Partial molar volumes at infinite dilution, V_2^∞, of tetra-*n*-alkylammonium bromides, R_4NBr (R = methyl, ethyl, propyl, butyl, pentyl), have been determined in binary mixtures of water with *N*,*N*-dimethylformamide (DMF) over the entire composition range at 298.15 K. Variations of V_2^∞ with the mole fraction of DMF as a function of solvent composition and electrolyte are considered. A linear dependence between V_2^∞ of the electrolyte and the molecular weight of the tetraalkylammonium cation was found.

Introduction

This work is part of a systematic study of limiting partial molar volumes of electrolytes, V_2^∞, in binary mixtures of water with several dipolar aprotic solvents. In a previous paper (García-Pañeda et al., 1994) V_2^∞ of alkali-metal halides (MX), Ph_4PCl, and $NaPh_4B$ in aqueous mixtures of *N*,*N*-dimethylformamide (DMF), covering the entire mole fraction range, were reported. The volumetric behavior exhibited by MX revealed a regular decrease of limiting volume with the increasing composition of DMF. Nevertheless, the V_2^∞ of Ph_4PCl and $NaPh_4B$ showed a strong dependence on the DMF composition, with a maximum in the water rich region.

In the present work, limiting partial molar volumes of tetra-*n*-alkylammonium bromides in aqueous mixtures with DMF are reported over the entire range of mole fraction at 298.15 K.

Experimental Section

Materials. *N*,*N*-Dimethylformamide (Merck, stated purity 99.8%, maximum content H_2O <0.05%) was dried for several days over a thermally activated 4A molecular sieve prior to use. Nonane was from Fluka (74252, stated purity >99%). Tetra-*n*-alkylammonium bromides, R_4NBr (R = methyl, ethyl, propyl, butyl, pentyl), from Fluka or Merck of the best quality available, were purified and checked as described by Conway et al. (1966). They were kept in a vacuum desiccator with P_2O_5 prior to use. Water from a Milli-Ro and Milli-Q water system (Millipore, $\kappa \approx 10^{-6}$ S·cm^{-1}) was used for preparation of solutions. Solvent and electrolyte solutions were prepared by mass using a Mettler AE 160 balance with an accuracy of ±0.0001 g, and the accuracy of the calculations of the molality was estimated at ±0.0001 mol·kg^{-1}.

Measurements. The solution densities were measured at 298.15 K using a vibrating-tube densitometer (A. Paar, DMA 602). The required temperature constancy of ±0.01 K was achieved using a cascade water bath arrangement (Heto DBT connected to Hetofrig cooling bath CB7). The temperature control was monitored to ±0.001 K inside the cell near the vibrating tube with a platinum resistance

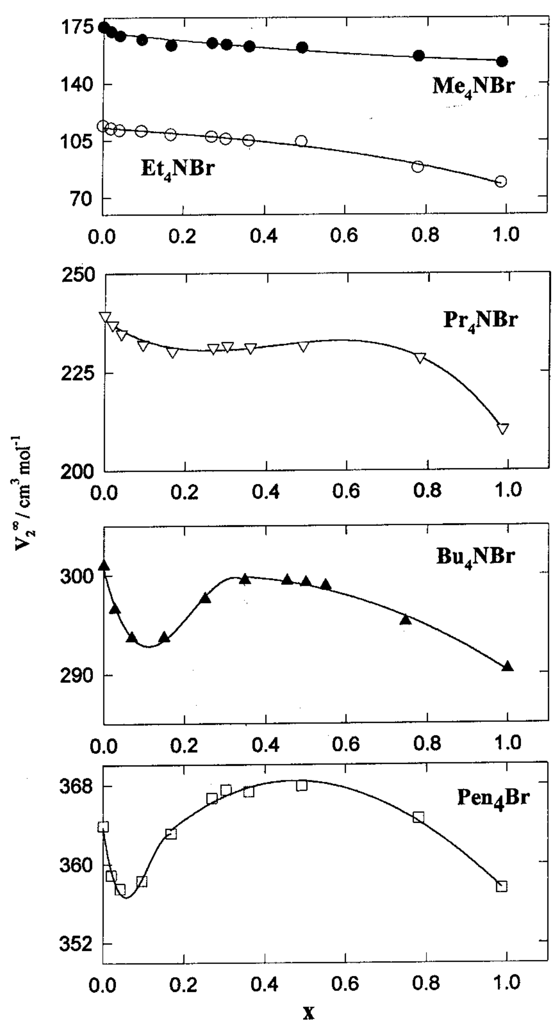

Figure 1. Variation of limiting partial molar volumes, V_2^∞, of tetraalkylammonium bromides with DMF mole fraction, *x*, at 298.15 K: (—) polynomial regression.

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Table 1. Molalities (m), Densities (ρ), and Apparent Molar Volumes (V_ϕ) for Tetra-*n*-alkylammonium Bromides in xDMF + (1 − x)Water Mixures at 298.15 K

m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}
				x = 0.0196				
				Me_4NBr				
0.0	0.996 251		0.1334	1.001 651	113.18	0.2044	1.004 412	113.42
0.0461	0.998 145	112.97	0.1412	1.001 951	113.26	0.2115	1.004 660	113.56
0.0770	0.999 395	113.08	0.1757	1.003 300	113.36	0.2386	1.005 731	113.47
0.1001	1.000 330	113.06	0.1791	1.003 420	113.43			
				Et_4NBr				
0.0485	0.998 141	171.36	0.1243	1.001 031	171.38	0.1835	1.003 221	171.48
0.0712	0.999 012	171.41	0.1425	1.001 711	171.41			
0.1043	1.000 260	171.53	0.1656	1.002 870	171.42			
				Pr_4NBr				
0.0506	0.997 801	236.44	0.1188	0.999 937	235.54	0.1864	1.001 967	235.43
0.0683	0.998 364	236.00	0.1414	1.000 592	235.71	0.2175	1.002 906	235.27
0.0961	0.999 216	235.89	0.1697	1.001 461	235.51	0.2213	1.003 023	235.24
				Bu_4NBr				
0.0488	0.997 553	296.22	0.1140	0.999 262	295.99	0.1979	1.001 420	295.64
0.0689	0.998 091	296.04	0.1366	0.999 851	295.87	0.2092	1.001 700	295.63
0.0973	0.998 840	295.91	0.1588	1.000 420	295.80	0.2210	1.002 050	295.34
				Pen_4NBr				
0.1215	0.998 650	359.16	0.1702	0.999 552	359.18	0.2292	1.000 602	359.22
0.1510	0.999 201	359.17	0.2010	1.000 110	359.18	0.2365	1.000 727	359.23
				x = 0.0417				
				Me_4NBr				
0.0	0.996 270		0.1225	1.001 349	112.29	0.2350	1.005 823	112.60
0.0488	0.998 329	111.90	0.1458	1.002 276	112.46	0.2499	1.006 385	112.71
0.0773	0.999 505	112.11	0.1710	1.003 258	112.67			
0.1027	1.000 549	112.18	0.1863	1.003 903	112.50			
				Et_4NBr				
0.0463	0.998 178	169.10	0.1308	1.001 535	169.50	0.1968	1.004 104	169.51
0.0717	0.999 200	169.28	0.1475	1.002 195	169.47	0.2271	1.005 263	169.52
0.1090	1.000 683	169.41	0.1718	1.003 136	169.51	0.2519	1.006 232	169.41
				Pr_4NBr				
0.0554	0.998 090	234.14	0.1145	1.000 030	233.70	0.1965	1.002 671	233.35
0.0747	0.998 732	233.88	0.1516	1.001 233	233.52	0.2147	1.003 250	233.28
0.0914	0.999 283	233.75	0.1733	1.001 920	233.50	0.2467	1.004 260	233.17
				Bu_4NBr				
0.0493	0.997 679	294.28	0.1449	1.000 312	294.19	0.2221	1.002 375	293.99
0.0978	0.999 021	294.33	0.1673	1.000 923	294.09	0.2317	1.002 641	293.90
0.1166	0.999 541	294.26	0.1894	1.001 520	294.01			
				Pen_4NBr				
0.0474	0.997 280	358.08	0.1208	0.998 775	358.11	0.1949	1.000 170	358.34
0.0667	0.997 684	358.04	0.1480	0.999 313	358.10	0.2247	1.000 686	358.52
0.0948	0.998 261	358.03	0.1739	0.999 780	358.31	0.2479	1.001 091	358.58
				x = 0.0995				
				Me_4NBr				
0.0	0.996 919		0.1340	1.002 609	111.18	0.2299	1.006 571	111.22
0.0617	0.999 560	111.17	0.1451	1.003 077	111.15	0.2351	1.006 738	111.42
0.0807	1.000 379	111.01	0.1921	1.005 024	111.18			
0.1053	1.001 415	111.07	0.2066	1.005 631	111.14			
				Et_4NBr				
0.0473	0.998 963	166.99	0.1325	1.002 555	167.07	0.2320	1.006 623	167.10
0.0594	0.999 491	166.81	0.1487	1.003 224	167.09	0.2501	1.007 355	167.07
0.0704	0.999 959	166.85	0.1716	1.004 163	167.12			
0.1011	1.001 251	166.97	0.2004	1.005 342	167.11			
				Pr_4NBr				
0.0526	0.998 763	231.39	0.1233	1.001 230	230.91	0.1990	1.003 855	230.42
0.0736	0.999 512	231.04	0.1515	1.002 207	230.75	0.2221	1.004 620	230.42
0.0927	1.000 181	230.93	0.1800	1.003 226	230.38	0.2521	1.005 611	230.39
				Bu_4NBr				
0.0485	0.998 361	292.94	0.1204	1.000 400	293.16	0.1993	1.002 450	
0.0668	0.998 891	292.99	0.1393	1.000 900	293.35	0.2124	1.002 822	
0.0751	0.999 130	293.00	0.1783	1.001 962		0.2375	1.003 500	
0.0876	0.999 450	293.46	0.1940	1.002 341				
				Pen_4NBr				
0.0452	0.997 812	359.46	0.1205	0.999 210	359.70	0.1979	1.000 492	360.20
0.0697	0.998 286	359.43	0.1472	0.999 683	359.77	0.2125	1.000 724	360.27
0.0980	0.998 801	359.66	0.1756	1.000 130	360.10	0.2286	1.000 887	360.76
				x = 0.1678				
				Me_4NBr				
0.0	0.996 775		0.1294	1.002 473	109.61	0.2158	1.006 122	109.93
0.0454	0.998 820	109.30	0.1612	1.003 823	109.78	0.2480	1.007 463	110.00
0.0777	1.000 233	109.39	0.1698	1.004 188	109.80			
0.1048	1.001 403	109.60	0.2117	1.005 952	109.92			

Table 1 (Continued)

m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}
				Et_4NBr				
0.0497	0.999 069	164.01	0.1261	1.002 412	164.91	0.2144	1.006 146	165.29
0.0710	1.000 007	164.49	0.1527	1.003 550	165.06	0.2394	1.007 164	165.44
0.0967	1.001 130	164.79	0.1729	1.004 408	165.14			
				Pr_4NBr				
0.0747	0.999 480	230.46	0.1469	1.001 991	230.59	0.2265	1.004 650	230.72
0.0905	1.000 020	230.69	0.1766	1.002 972	230.78			
0.1164	1.000 950	230.47	0.1923	1.003 530	230.61			
				Bu_4NBr				
0.0462	0.998 065	294.84	0.1105	0.999 723	295.60	0.1924	1.001 832	295.37
0.0726	0.998 752	295.33	0.1466	1.000 610	295.86	0.1962	1.001 920	295.40
0.0957	0.999 321	295.79	0.1694	1.001 263	295.33	0.2477	1.003 131	295.61
				Pen_4NBr				
0.0449	0.997 471	363.83	0.1291	0.998 630	364.55	0.1987	0.999 601	364.21
0.0700	0.997 859	363.71	0.1347	0.998 738	364.31			
0.1047	0.998 360	363.87	0.1737	0.999 289	364.55			
				$x = 0.2681$				
				Me_4NBr				
0.0	0.992 982		0.1304	0.998 896	108.51	0.2313	1.003 307	108.75
0.0534	0.995 449	108.03	0.1601	1.000 231	108.44	0.2514	1.004 180	108.75
0.0679	0.996 122	107.91	0.1878	1.001 397	108.78			
0.0901	0.997 062	108.78	0.1975	1.001 852	108.63			
				Et_4NBr				
0.0503	0.995 289	164.75	0.1531	0.999 861	164.93	0.2167	1.002 615	164.96
0.1018	0.997 601	164.86	0.1748	1.000 797	165.00			
0.1333	0.998 998	164.87	0.2047	1.002 108	164.91			
				Pr_4NBr				
0.0516	0.994 855	231.31	0.1291	0.997 525	231.81	0.2161	1.000 410	231.96
0.0679	0.995 414	231.66	0.1568	0.998 495	231.61	0.2471	1.001 408	232.00
0.0989	0.996 496	231.70	0.1883	0.999 569	231.54	0.2869	1.002 767	231.68
				Bu_4NBr				
0.0516	0.994 322	297.01	0.1237	0.996 042	298.55	0.1893	0.997 571	298.59
0.0677	0.994 720	298.00	0.1472	0.996 610	298.47	0.2088	0.997 990	298.73
0.0949	0.995 391	298.09	0.1606	0.996 900	298.64	0.2346	0.998 553	298.80
				Pen_4NBr				
0.0438	0.993 561	367.54	0.1274	0.994 586	367.80	0.2106	0.995 511	368.05
0.0697	0.993 892	367.59	0.1556	0.994 891	368.01	0.2491	0.995 828	368.52
0.0983	0.994 250	367.61	0.1931	0.995 279	368.25			
				$x = 0.3028$				
				Me_4NBr				
0.0	0.990 297		0.1249	0.996 055	107.93	0.2358	1.000 919	108.47
0.0559	0.992 927	107.31	0.1709	0.998 071	108.33	0.2631	1.002 084	108.59
0.0763	0.993 897	107.07	0.2075	0.999 667	108.50			
				Et_4NBr				
0.0705	0.993 575	164.26	0.1815	0.998 446	165.08	0.2691	1.002 244	164.96
0.0883	0.994 381	164.38	0.2068	0.999 605	164.77			
0.1299	0.996 243	164.56	0.2418	1.001 119	164.78			
				Pr_4NBr				
0.0534	0.992 215	231.81	0.1227	0.994 601	232.10	0.2175	0.997 694	232.46
0.0709	0.992 801	232.28	0.1454	0.995 306	232.57	0.2476	0.998 701	232.30
0.1088	0.994 064	232.69	0.1899	0.996 783	232.53	0.2558	0.998 839	232.82
				Pen_4NBr				
0.0446	0.990 876	368.75	0.1046	0.991 560	369.41	0.2151	0.992 782	369.49
0.0542	0.990 986	368.98	0.1378	0.991 960	369.27	0.2331	0.992 961	369.55
0.0731	0.991 222	368.95	0.1853	0.992 475	369.40	0.2686	0.993 306	369.65
				$x = 0.3594$				
				Me_4NBr				
0.0	0.986 636		0.1098	0.991 864	106.67	0.1658	0.994 398	107.21
0.0504	0.989 073	106.21	0.1306	0.992 848	106.61	0.2066	0.996 251	107.29
0.0807	0.990 509	106.43	0.1404	0.993 208	107.35	0.2381	0.997 639	107.48
				$x = 0.3594$				
				Et_4NBr				
0.0672	0.989 847	163.39	0.1727	0.994 624	164.16	0.2352	0.997 357	164.39
0.0890	0.990 857	163.59	0.1942	0.995 569	164.27	0.2412	0.997 615	164.42
0.1346	0.992 899	164.17	0.2130	0.996 396	164.31			
				Pr_4NBr				
0.0540	0.988 625	231.57	0.1221	0.991 021	231.95	0.2088	0.997 694	232.46
0.0697	0.989 185	231.71	0.1508	0.992 019	231.94	0.2451	0.998 701	232.30
0.0985	0.990 204	231.83	0.1865	0.996 783	232.53	0.2563	0.998 839	232.82
				Pen_4Br				
0.0451	0.987 296	368.34	0.1263	0.988 314	369.34	0.1894	0.989 053	369.60
0.0901	0.987 879	368.98	0.1596	0.988 715	369.46			
0.0732	0.987 660	368.86	0.1858	0.989 008	369.61			

Table 1 (Continued)

m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}	m/mol·kg^{-1}	ρ/g·cm^{-3}	V_ϕ/cm^3·mol^{-1}
				$x = 0.4903$				
				Me_4NBr				
0.0	0.976 444		0.1079	0.981 724	105.88	0.2052	0.986 273	106.47
0.0467	0.978 766	105.38	0.1575	0.984 092	106.02	0.2372	0.987 763	106.49
0.0762	0.980 212	105.51	0.1858	0.985 431	106.07			
				Et_4NBr				
0.0698	0.979 900	162.72	0.1809	0.985 164	163.22	0.2270	0.987 273	163.38
0.1022	0.981 457	162.95	0.1984	0.985 966	163.30	0.2572	0.988 629	163.50
0.1495	0.983 749	162.76	0.2219	0.987 047	163.34			
				$x = 0.4903$				
				Pr_4NBr				
0.0744	0.979 298	232.19	0.1895	0.983 475	232.51	0.2396	0.985 204	232.67
0.1336	0.981 480	232.37	0.2077	0.984 127	232.48			
0.1601	0.982 429	232.47	0.2169	0.984 441	232.63			
				Pen_4NBr				
0.0459	0.977 240	369.13	0.1178	0.978 387	369.59	0.1968	0.979 594	369.64
0.0533	0.977 407	368.31	0.1607	0.979 038	369.71	0.2292	0.980 017	369.92
0.0773	0.977 753	369.36	0.1797	0.979 330	369.68	0.2444	0.980 210	370.03
				$x = 0.7793$				
				Et_4NBr				
0.0	0.956 911		0.1565	0.965 362	159.24	0.2819	0.971 715	159.80
0.0524	0.959 855	157.78	0.1768	0.966 447	159.13	0.3227	0.973 641	160.20
0.0708	0.960 868	157.93	0.2151	0.968 356	159.61			
0.0962	0.962 211	158.58	0.2538	0.970 304	159.76			
				Pr_4NBr				
0.0553	0.959 376	228.99	0.1208	0.962 242	228.79	0.2103	0.965 959	229.11
0.0694	0.960 017	228.64	0.1596	0.963 868	228.99	0.2281	0.966 702	229.04
0.0944	0.961 081	229.02	0.1822	0.964 822	228.95			
				Pen_4NBr				
0.0444	0.958 113	365.51	0.1265	0.960 198	365.90	0.2467	0.962 888	366.78
0.0727	0.958 851	365.65	0.1605	0.960 976	366.32	0.2712	0.963453	366.68
0.0947	0.959 392	365.97	0.1923	0.961 681	366.62			
				$x = 0.9858$				
				Et_4NBr				
0.0	0.944 139		0.1365	0.952 305	154.15	0.2631	0.959 493	154.61
0.0517	0.947 304	153.40	0.1548	0.953 277	154.87	0.3050	0.961 630	155.38
0.0722	0.948 529	153.67	0.2114	0.956 488	155.03			
0.0966	0.950 131	154.13	0.2331	0.957 816	154.53			
				Pr_4NBr				
0.0535	0.947 163	217.92	0.1189	0.950 473	220.78	0.1909	0.953 474	224.94
0.0666	0.947 883	218.09	0.1575	0.952 241	222.41	0.2195	0.954 751	225.26
0.0928	0.949 159	220.17	0.1806	0.983 046	224.58	0.2775	0.957 151	226.30
				Pen_4Br				
0.0453	0.945 779	359.65	0.1307	0.948 583	361.04	0.2241	0.951 440	361.54
0.0717	0.946 691	359.98	0.1585	0.949 432	361.39	0.2565	0.952 273	362.19
0.0976	0.947 585	359.96	0.1901	0.950 371	361.72	0.2643	0.952 415	362.58

thermometer connected to an ASL model F250 resistance bridge, which was calibrated by Isotech (England) with a precision of ±0.001 K and accuracy in absolute temperature of ±0.010 K (ITS-90). The whole system was placed in a room in which the temperature was kept constant within ±0.5 K. The output signal from the oscillating tube of the densitometer was processed through a frequency meter (A. Paar DMA 60). The density of the fluid in the measuring oscillating tube, ρ, was related to the oscillating period, τ, by the equation

$$\rho = A + B\tau^2 \qquad (1)$$

where A and B are instrument constants which can be determined by calibration with fluids of known density. Water (ρ = 0.997 045 g·cm^{-3}) (Kell, 1975) and nonane (ρ = 0.713 85 g·cm^{-3}) (Ortega and Paz-Andrade, 1986) were used as the calibration standards. In order to test the instrument, the partial molar volume at infinite dilution of NaCl in water was found to be 16.63 ± 0.15 cm^3·mol^{-1}, which is in agreement with the average (16.55 ± 0.08 cm^3·mol^{-1}) of the 29 values tabulated by Millero (1972). Densities of solvent mixtures and electrolyte solutions had an uncertainty (95% confidence limits) of ±7 × 10^{-6} g cm^{-3}.

Results and Discussion

The apparent molar volumes V_ϕ of n-R_4NBr in aqueous mixtures of DMF were calculated from the densities of the solutions using the standard expression:

$$V_\phi = M_2/\rho + (\rho_0 - \rho)/m\rho\rho_0 \qquad (2)$$

where M_2 is the molecular weight of the electrolyte, m is its molality, and ρ and ρ_0 represent the density of solution and solvent, respectively. The values of V_ϕ at various molalities (0 to 0.323) mol·kg^{-1}, along with the respective densities, are given in Table 1. The application of the Redlich−Meyer equation (1964) to obtain V_ϕ^∞ was not possible due to lack of the theoretical Debye−Hückel slopes, S_v, in aqueous mixtures of DMF. However, V_ϕ was found to vary linearly with $m^{1/2}$ over the concentration range investigated. The limiting partial molar volume of the tetraalkylammonium bromides, $V_2^\infty = V_\phi^\infty$, was obtained by least-squares fitting of the results to the Masson empirical equation (Millero, 1972)

$$V_\phi = V_\phi^\infty + S_v^* m^{1/2} \qquad (3)$$

where S_V^* is the experimental slope. Table 2 shows values of partial molar volumes at infinite dilution, V_2^∞, along with their 95% confidence limits (in parentheses), and results also given by other workers (Millero, 1972; Heuvelsland and Somsen, 1977; Dey et al., 1985). The low solubility of Me_4NBr at 0.7793 and 0.9858 mole fraction prevented us from obtaining measurements of density with the required precision to calculate the corresponding apparent molar volumes. Nevertheless, a good linear dependence between V_2^∞ of tetra-*n*-alkylammonium bromides and the molecular weight of the R_4N^+ cation, M_C, over the entire composition range was found. Plots of V_2^∞ of tetraalkylammonium bromides vs M_C were analyzed by a linear regression analysis and the following expressions obtained:

$$x = 0.0196; \quad V_2^\infty = 29.96 + 1.102 M_C; \quad \sigma(V_2^\infty) = 1.07 \quad (4)$$

$$x = 0.0417; \quad V_2^\infty = 28.12 + 1.102 M_C; \quad \sigma(V_2^\infty) = 1.75 \quad (5)$$

$$x = 0.0955; \quad V_2^\infty = 25.97 + 1.106 M_C; \quad \sigma(V_2^\infty) = 2.60 \quad (6)$$

$$x = 0.1678; \quad V_2^\infty = 19.23 + 1.142 M_C; \quad \sigma(V_2^\infty) = 3.86 \quad (7)$$

$$x = 0.2681; \quad V_2^\infty = 17.26 + 1.159 M_C; \quad \sigma(V_2^\infty) = 3.49 \quad (8)$$

$$x = 0.3028; \quad V_2^\infty = 14.44 + 1.176 M_C; \quad \sigma(V_2^\infty) = 3.39 \quad (9)$$

$$x = 0.3594; \quad V_2^\infty = 13.32 + 1.179 M_C; \quad \sigma(V_2^\infty) = 8.58 \quad (10)$$

$$x = 0.4903; \quad V_2^\infty = 12.28 + 1.185 M_C; \quad \sigma(V_2^\infty) = 3.41 \quad (11)$$

$$x = 0.7793; \quad V_2^\infty = -2.989 + 1.231 M_C; \quad \sigma(V_2^\infty) = 1.51 \quad (12)$$

$$x = 0.9858; \quad V_2^\infty = -13.06 + 1.244 M_C; \quad \sigma(V_2^\infty) = 5.25 \quad (13)$$

where x is the mole fraction of DMF and $\sigma(V_2^\infty)$ denotes the standard deviation. Limiting partial molar volumes of Me_4NBr at 0.7793 and 0.9858 mole fraction that appear in Table 2 were calculated from expressions 12 and 13, respectively.

Variations of V_2^∞ for R_4NBr with the DMF composition are shown in Figure 1. The effect of DMF on the limiting volume produces different trends which depend on the electrolyte and solvent compositions.

Table 2. Limiting Partial Molar Volumes, V_2^∞, for Tetra-*n*-alkylammonium Bromides in xDMF + (1 − x) Water Mixtures at 298.15 K

	V_2^∞/cm^3·mol^{-1}				
x	Me_4NBr	Et_4NBr	Pr_4NBr	Bu_4NBr	Pen_4NBr
0.0	114.2[a]	174.3[a]	239.6[a]	301.0[a]	363.9[a]
0.0196	112.5 (0.2)	171.3 (0.3)	237.2 (0.4)	296.8 (0.3)	358.9 (0.1)
0.0282				296.6[b]	
0.0417	111.3 (0.3)	168.9 (0.3)	234.9 (0.2)	294.7 (0.3)	357.5 (0.3)
0.0580	111[c]	186[c]		291[c]	
0.0700				293.7[b]	
0.0955	110.9 (0.3)	166.7 (0.2)	232.1 (0.4)	292.4 (0.4)	358.3 (0.6)
0.1411	113[c]	170[c]	291[c]		
0.1500				293.7[b]	
0.1678	108.7 (0.1)	163.1 (0.4)	230.3 (0.6)	294.9 (0.9)	363.1 (0.9)
0.2503				297.6[b]	
0.2681	107.5 (0.8)	164.6 (0.2)	231.2 (0.6)	297.0 (0.3)	366.7 (0.5)
0.2699	121[c]	171[c]			
0.3028	105.9 (0.7)	163.6 (0.7)	231.7 (0.8)	303[c]	368.3 (0.3)
0.3482				299.6[b]	
0.3594	105.0 (0.7)	162.4 (0.5)	231.3 (0.2)		367.3 (0.4)
0.4529				299.4[b]	
0.4903	104.4 (0.4)	161.8 (0.6)	231.7 (0.3)		367.9 (0.8)
0.4964	108[c]	166[c]		300[c]	
0.4996				299.2[b]	
0.5491				298.9[b]	
0.7462				295.2[b]	
0.7793	88.3[d]	156.3 (0.4)	228.6 (0.5)		364.6 (0.4)
0.9858	78.9[d]	152.4 (0.9)	210.4 (1.0)		257.5 (0.8)
1.0		148.4[e]	220.9[e]	290.4[b]	359.6[e]
				289.3[e]	

[a] Millero (1972). [b] Heuvelsland and Somsen (1977). [c] Dey et al. (1985). [d] Calculated from expressions 12 and 13. [e] Kawaizumi and Zana (1974). Values in parentheses are 95% confidence limits.

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