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Apparent Molar Volumes of Some Complex Cyanides in Aqueous Solutions at 15–60 °C

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The apparent molar volumes (ϕ_V) of a series of six complex cyanide compounds have been measured in aqueous solutions in the concentration range 0.001–0.5 mol dm^{-3}, and at five temperatures in the range 15–60 °C. The compounds studied are $K[Ag(CN)_2]$, $K_2[Ni(CN)_4]$, $K_3[Co(CN)_6]$, $K_3[Fe(CN)_6]$, $K_4[Fe(CN)_6]$, and $K_4[Mo(CN)_8]$. At higher concentrations ($c > 0.05$ mol dm^{-3}) ϕ_V was determined directly from pycnometer density measurements, while in the lower concentration region the dilatometer technique of Hepler, Stokes, and Stokes was employed.

The apparent molar volumes (ϕ_V) of many simple salts and salts containing large hydrophobic ions have been accurately measured in aqueous solutions (cf. the extensive compilation of Millero (*12*)). Few data, however, are available for the molar volumes of large hydrophilic ions or complex ions. Therefore we have measured apparent molar volumes as part of an extended study (*9, 10*) of the thermodynamic and transport properties of aqueous solutions of some complex cyanide compounds. In this paper we report ϕ_V measurements for $K[Ag(CN)_2]$, $K_2[Ni(CN)_4]$, $K_3[Co(CN)_6]$, $K_3[Fe(CN)_6]$, $K_4[Fe(CN)_6]$, and $K_4[Mo(CN)_8]$. Studies were made at 15, 25, 35, 45, and 60 °C using pycnometric density determinations, accurate to $\pm 1 \times 10^{-5}$ g cm^{-3}, for solutions of concentration $0.5 > c > 0.05$ mol dm^{-3}. For solutions of lower concentration pycnometry does not yield sufficiently accurate ϕ_V values (see below) and the dilatometer technique of Hepler, Stokes, and Stokes (*6*) was employed in the concentration region $0.001 < c < 0.05$ mol dm^{-1}. Infinite dilution values of ϕ_V (i.e., $\phi_V{}^0$) have been determined by an extrapolation procedure which assumes Debye-Hückel limiting law behavior at low concentrations.

Experimental Section

Density measurements were made according to recommended methods (*15*) using two Sprengel-Ostwald pycnometers of approximately 30-cm^3 capacity with arms made of capillary tubing of 0.3 mm internal diameter. Before weighing, the filled pycnometers were equilibrated for about an hour in a large (600 L) efficiently stirred, insulated bath, whose temperature was controlled to $\pm 5 \times 10^{-4}$ °C or better by a mercury-toluene regulator operating a 60-W light bulb as the on-off heater. Buoyancy corrections were made and duplicate measurements agreed to $\pm 1 \times 10^{-5}$ g cm^{-3}. The bath temperature was calibrated against standardized mercury in glass thermometers. This bath was also used for the dilatometer measurements.

Dilatometer measurement techniques essentially similar to those described by Hepler, Stokes, and Stokes (*6*) were employed in this study. Dilatometers from 300 cm^3 to 1 dm^3 with capillaries of 1.00 mm diameter and with capsules from 3 to 60 cm^3 provided an adequate range of final concentrations. The measurement consists of observing the change in height (corrected for hydrostatic head) of solution in the capillary (Δh(corr)) as concentrated solution in the capsule, of known ϕ_V from density measurement, is mixed with pure solvent in the dilatometer flask. Since the capillary diameter is known Δh(corr) can be directly related to the volume change on dilution, Δv.

Calculations and Errors

The apparent molar volume of a solute in a solution of density d, g cm^{-3}, is given by,

$$\phi_V = \bar{M}_2/d_0 - 1000(d - d_0)/cd_0 \ldots \qquad (1)$$

where $\bar{M}_2$ is the molecular weight of the solute, d_0 is the solvent density, and c is the solute concentration in mol dm^{-3}. Errors in the concentration of carefully prepared solutions do not affect ϕ_V significantly; however, differentiation of eq 1 at constant concentration gives,

$$\delta\phi_V = \left|\frac{1000}{c}\frac{\delta d}{d}\right| \qquad (2)$$

Thus for a given uncertainty in density, the uncertainty in ϕ_V is inversely proportional to concentration. Using an uncertainty in density appropriate for our pycnometers, i.e., $\delta d/d = 2 \times 10^{-5}$ $\delta\phi_V$ is already 0.2 cm^3 mol^{-1} at 0.1 mol dm^{-3} and increases rapidly at lower concentrations.

In dilatometer experiments the apparent molar volume of the final dilute solution $\phi_V{}^{final}$ is related to that of the initial concentrated solution, $\phi_V{}^{init}$ (determined by density measurement)

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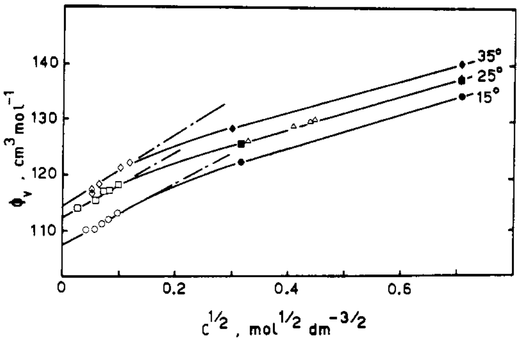

Figure 1. ϕ_V as a function of $c^{1/2}$ for $K_4[Fe(CN)_6]$ at 15, 25, and 35 °C. Triangles represent data of Hepler, Stokes, and Stokes (*4*). Broken lines are Debye–Hückel limiting slopes.

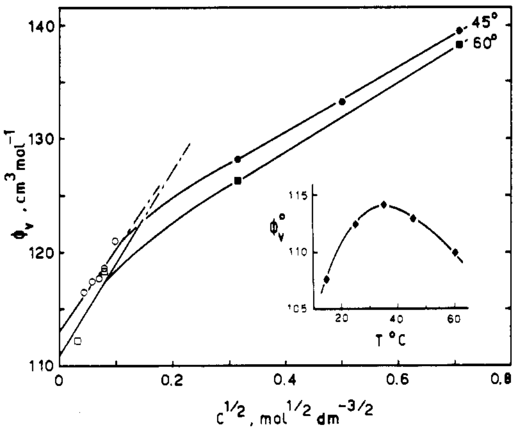

Figure 2. ϕ_V as a function of $c^{1/2}$ for $K_4[Fe(CN)_6]$ at 45 and 60 °C. Extrapolated ϕ_V^0 values are shown as a function of temperature.

by,

$$\phi_V^{\mathrm{final}} = \phi_V^{\mathrm{init}} + \Delta v/n_2 \qquad (3)$$

where n_2 is the number of moles of solute involved in the dilatometer experiment. The error in ϕ_V^{final} is thus the error in ϕ_V^{init} plus that in $\Delta v/n_2$. Since solutions are made up analytically and capsule volumes are accurately calibrated the error in n_2 contributes negligibly to that in ϕ_V^{final}. The error in Δv is due to the uncertainty in Δh(corr) (ca. ±0.003 cm); this is the approximate reproducibility in cathetometer measurements of Δh(corr) and includes the influence of temperature variations of $\pm 5 \times 10^{-4}$ °C. To this we add an uncertainty volume change on opening the capsule, of say, half the uncertainty in Δh(corr). $\delta\Delta v$ is thus ca. 7×10^{-5} cm^3. For a typical capsule of volume 7 cm^3, $n_2 = 7 \times 10^{-3} c$ mol and,

$$\delta\phi_V^{\mathrm{final}} = \delta\phi_V^{\mathrm{init}} + \frac{10^{-2}}{c} \qquad (4)$$

If the initial concentration is say 1 mol dm^{-3}, $\delta\phi_V^{\mathrm{final}} = 0.02$ cm^3 mol^{-1}, typical of the accuracy achieved and claimed by Hepler, Stokes, and Stokes (*6*), and apparently accepted for all situations by others. However, for solutions of 0.1 mol dm^{-3}, typical for many of our experiments, $\delta\phi_V^{\mathrm{final}}$ rises to $\delta\phi_V^{\mathrm{init}} + 0.1$ cm^3 mol^{-1}, or more if smaller capsules are employed to achieve lower final concentrations. Of course the dilatometer method of ϕ_V determination is much more accurate than typical pycnometer density determination for solutions of low concentration.

Materials

$K_3[Fe(CN)_6]$ and $K_4[Fe(CN)_6]$ were purified by recrystallization of the A.R. grade compounds from conductance water. They were dried overnight at 110 °C, (the Fe(II) compound losing its water of crystallization), and then stored in a vacuum desiccator over P_2O_5. The Fe content was estimated gravimetrically as the oxinate.[6] $K_3[Co(CN)_6]$ was prepared by the method of Brigelow (*2*) and the Co content was determined by EDTA titration using pyrocatechol violet as indicator (*18*). $K_4[Mo(CN)_8]\cdot 2H_2O$ was prepared by the method of Furman and Miller (*5*) and the metal content was analyzed by titration of the complex with Ce(IV) using *o*-phenanthrolineiron(II) indicator (*5*). $K_2[Ni(CN)_4]$ was prepared by the method of Fernelius and Burbage (*4*), but maintaining excess insoluble $Ni(CN)_2$ as the complex is formed after further addition of KCN. This procedure eliminates the production of five-coordinate species. Water of crystallization was removed by heating in an oven overnight at 120 °C. Ni was analyzed by EDTA titration using murexide as indicator (*16*). $K[Ag(CN)_2]$ was prepared by precipitation of AgCN from an equimolar solution of $AgNO_3$ and KCN. The AgCN was dissolved by further addition of KCN. The formation of higher complexes was avoided by maintaining excess AgCN in the solution which was filtered, reduced in volume on the hot plate, and set aside for crystallization. The complex was recrystallized from conductance water and was dried and stored over P_2O_5 in a vacuum desiccator. Ag was determined gravimetrically as the chloride (*17*). Each of the complexes was analyzed for C and N content by the Australian Microanalytical Service. Analyses and purities (based on metal analysis) are shown in Table I.

Water used in the preparation of solutions and in the dilatometer experiments was distilled from an all-glass apparatus and then passed through a mixed bed ion-exchange column and

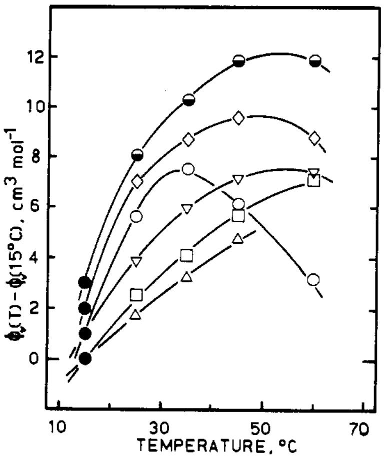

Figure 3. Temperature variation of ϕ_V^0. $\phi_V^0(T) - \phi_V^0(15\ °C)$ is shown as a function of temperature: ●, $K_3[Fe(CN)_6]$; ◇, $K_4[Mo(CN)_8]$; ○, $K_4[Fe(CN)_6]$; ▽, $K_3[Co(CN)_6]$; □, $K[Ag(CN)_2]$; △, $K_2[Ni(CN)_4]$. Zeros have been offset in some cases for clarity.

Table I. Analyses of Cyano Complexes

		% C	% N	% metal	% purity
$K[Ag(CN)_2]$	Theor	12.0	14.2	54.23	
	Found	11.8	14.2	54.10	99.8
$K_2[Ni(CN)_4]$	Theor	19.9	22.4	24.36	
	Found	19.4	21.8	24.25	99.5
$K_3[Co(CN)_6]$	Theor	21.7	25.3	17.73	
	Found	21.6	25.3	17.70	99.8
$K_3[Fe(CN)_6]$	Theor	21.9	25.5	16.96	
	Found	21.8	25.3	16.95	100.0
$K_4[Fe(CN)_6]$	Theor	19.5	22.8	15.16	
	Found	19.6	22.8	15.14	99.9
$K_4[Mo(CN)_8]\cdot 2H_2O$	Theor	19.3	22.5	19.32	
	Found	19.1	22.3	19.36	100.2

Table II

Temp, °C	Concn, mol dm^{-3}	ϕ_V, $cm^3\ mol^{-1}$	Temp, °C	Concn, mol dm^{-3}	ϕ_V, $cm^3\ mol^{-1}$
	$K[Ag(CN)_2]$			$K_2[Ni(CN)_4]$	
15	*0.500*	*71.13*	15	*0.200*	*114.89*
	0.250	*70.11*		0.020 77	112.83
	0.009 85	69.59		0.005 67	112.38
	0.003 50	69.43		0.002 01	111.97
	0.002 69	69.37		0.001 55	111.65
25	*0.500*	*73.19*	25	*0.200*	*116.48*
	0.250	*72.66*		*0.100*	*115.73*
	0.009 85	71.88		0.020 77	114.68
	0.003 50	71.78		0.005 67	113.90
	0.002 69	71.89		0.002 01	113.77
	0.001 76	71.98		0.002 01	113.58
35	*0.500*	*74.88*		0.001 55	113.59
	0.250	*74.24*		0.001 01	113.59
	0.009 85	73.70	35	*0.200*	*118.49*
	0.006 61	73.54		*0.100*	*117.72*
	0.003 50	73.38		0.020 77	116.26
	0.001 76	73.45		0.005 67	115.68
45	*0.500*	*76.10*		0.002 01	115.29
	0.250	*75.86*		0.002 01	115.32
	0.051 93	75.59		0.001 01	115.14
	0.006 61	75.18	45	*0.200*	*120.97*
	0.005 08	75.20		*0.100*	*120.46*
	0.001 76	75.11		0.020 77	118.14
60	*0.496*	*77.62*		0.005 67	117.51
	0.248	*77.68*		0.002 01	117.01
	0.006 56	76.58		0.001 55	116.82
	0.006 56	76.62			
	0.003 28	76.61			
	$K_4[Fe(CN)_6]$			$K_4[Mo(CN)_8]$	
15	*0.500*	*134.56*	15	*0.200*	*178.65*
	0.100	*122.56*		*0.100*	*174.56*
	0.009 85	113.29		*0.050*	*170.46*
	0.006 62	112.14		0.010 39	165.21
	0.005 08	111.25		0.002 84	162.75
	0.003 32	110.11		0.001 00	161.73
	0.001 76	110.10		0.000 78	161.74
25	*0.500*	*137.88*	25	*0.200*	*184.20*
	0.100	*125.83*		*0.100*	*180.13*
	0.009 85	118.23		*0.050*	*176.62*
	0.000 70	114.15		0.010 39	171.34
	0.006 62	117.09		0.002 84	169.12
	0.005 08	117.18		0.001 00	167.52
	0.003 32	115.31		0.000 78	167.55
35	*0.500*	*140.11*	35	*0.200*	*185.80*
	0.100	*128.60*		*0.100*	*180.87*
	0.014 17	122.31		*0.050*	*176.95*
	0.010 39	121.44		0.010 39	172.02
	0.003 88	118.44		0.002 84	170.03
	0.002 84	116.73		0.001 00	168.69
	0.002 53	117.29		0.000 78	168.41

Temp, °C	Concn, mol dm^{-3}	ϕ_V, $cm^3\ mol^{-1}$	Temp, °C	Concn, mol dm^{-3}	ϕ_V, $cm^3\ mol^{-1}$
	$K_4[Fe(CN)_6]$			$K_4[Mo(CN)_8]$	
45	*0.500*	*139.44*	45	*0.200*	*189.64*
	0.250	*133.16*		*0.100*	*183.99*
	0.100	*128.33*		*0.050*	*179.85*
	0.009 85	121.03		0.002 84	172.70
	0.006 61	118.49		0.002 84	172.28
	0.005 08	117.68		0.001 00	169.84
	0.003 32	117.47		0.000 51	169.53
	0.001 76	116.49			
60	*0.500*	*138.24*	60	*0.092 7*	*183.33*
	0.100	*126.27*		0.001 22	170.00
	0.006 61	118.47		0.001 22	170.08
	0.001 32	112.07			
	$K_3[Co(CN)_6]$			$K_3[Fe(CN)_6]$	
15	*0.200*	*148.28*	15	*0.500*	*155.01*
	0.100	*145.88*		*0.100*	*148.79*
	0.020 77	143.30		0.051 93	146.42
	0.003 94	141.37		0.009 85	143.47
	0.002 65	140.72		0.006 62	142.87
	0.002 00	140.77		0.005 08	142.55
	0.001 33	139.84		0.003 32	142.47
	0.000 70	140.64		0.001 76	141.97
	$K_3[Co(CN)_6]$			$K_3[Fe(CN)_6]$	
25	*0.200*	*151.25*	25	*0.500*	*158.00*
	0.100	*149.30*		*0.100*	*152.30*
	0.020 78	146.63		0.009 85	148.77
	0.002 65	143.76		0.006 62	148.40
	0.002 03	143.02		0.005 08	148.11
	0.001 41	143.64		0.003 32	148.77
	0.001 32	143.31		0.001 41	146.79
35	*0.200*	*153.26*	35	*0.500*	*160.13*
	0.100	*151.64*		*0.100*	*154.88*
	0.020 77	148.80		0.014 17	151.59
	0.003 94	146.92		0.005 02	150.62
	0.002 65	145.34		0.003 88	150.11
	0.001 40	145.53		0.002 53	149.74
	0.000 70	144.93			
45	*0.200*	*154.71*	45	*0.500*	*161.83*
	0.100	*152.70*		*0.100*	*157.01*
	0.003 94	148.22		0.009 85	153.62
	0.002 64	147.04		0.006 61	152.71
	0.001 40	147.39		0.005 08	151.48
	0.001 40	146.94		0.003 32	151.87
	0.001 32	147.78		0.001 76	151.74
	0.000 70	146.68			
60	*0.200*	*155.86*	60	*0.500*	*162.29*
	0.100	*153.20*		*0.100*	*156.67*
	0.002 64	148.96		0.006 61	152.99
	0.002 64	148.58		0.006 61	152.69
	0.001 32	146.89		0.001 32	151.37

[a] Entries in italics represent pycnometer measurements.

stored polythene tanks. The conductivity ranged from 4 to 9 × $10^{-7}\ \Omega^{-1}\ cm^{-1}$. All stock solutions were prepared in calibrated 250-cm^3 volumetric flasks and were used within a few days. They were protected from the action of light as much as possible.

Results and Discussion

The ϕ_V results are shown below in Table II.

At concentrations approaching infinite dilution ϕ_V should become a linear function of $c^{1/2}$ in accordance with the predictions of the Debye-Hückel limiting law. This we observed for all the compounds studied ($0.001 < c < 0.01$ mol dm^{-3}, i.e., $0.03 < c^{1/2} < 0.01$ $mol^{1/2}\ dm^{-3/2}$). Some typical examples of the ϕ_V vs. $c^{1/2}$ plots are shown in Figures 1 and 2 for $K_4[Fe(CN)_6]$. Debye-Hückel limiting behavior is shown as dotted lines joining the observed ϕ_V curves at low concentrations. Extrapolations to $\phi_V{}^0$ have been achieved in this way.

The derived $\phi_V{}^0$ values (equal to the partial molar volume $\overline{V}_2{}^0$ at infinite dilution) are shown in Table III.

In Figure 3 we show the variation of $\phi_V{}^0$ with temperature for

Table III

Temp, °C	Apparent molar volume at infinite dilution ϕ_V^0 or $\bar{V}_2^0$ (cm^2 mol^{-1})					
	$K[Ag(CN)_2]$	$K_2[Ni(CN)_4]$	$K_3[Co(CN)_6]$	$K_3[Fe(CN)_6]$	$K_4[Fe(CN)_6]$	$K_4[Mo(CN)_8]$
15	69.3	111.6	139.6	141.1	107.7	160.2
25	71.8	113.3	142.5	146.2	112.3	165.2
35	73.4	114.8	144.6	148.4	114.2	166.9
45	75.0	116.4	145.8	150.0	112.7	167.8
60[a]	76.4	—	146.0	150.0	110.0	167.0
Estimated error	±0.1	±0.2	±0.3	±0.1	±0.2	±0.3

[a] Dilatometer measurements at 60 °C are experimentally more difficult and are associated with a greater uncertainty.

each of the six compounds. In each case a smooth variation (within the experimental error) of ϕ_V with temperature is found. Four of the six compounds studied show a maximum in ϕ_V at temperatures between 30 and 55 °C. This behavior is reminiscent of the behavior of the smaller 1:1 and 2:1 valence type electrolytes studied by Dunn (*3*). Maxima in ϕ_V have been related to ionic radius and change and have been discussed by several authors (cf. review of Millero (*12*)). Elsewhere (*9*, *11*) we have shown that the behavior of the complex cyanide ions is consistent with that of the simple small hydrophilic 1:1 and 2:1 valence type electrolytes.

In general, comparison of the present work with that of other workers is made difficult for two reasons: (a) The ϕ_V^0 values reported elsewhere may not be directly comparable because of different methods of extrapolation to infinite dilution. (b) ϕ_V values at other concentrations may not be comparable because of the lack of presentation of raw data.

Indelli and co-workers (*1*, *8*), also using the dilatometer method of Hepler, Stokes and Stokes (*6*) have measured ϕ_V to very low concentrations (ca. 10^{-4} mol dm^{-3}) for three complex cyanides at 25 °C. Indelli and Zamboni (*8*) have studied $K_4[Mo(CN)_8]$ while Billi, Malatesta, Zamboni, and Indelli (*1*) have studied $K_3[Fe(CN)_6]$ and $K_4[Fe(CN)_6]$. Debye-Hückel limiting law behavior was observed for $K_4[Mo(CN)_8]$ and $K_3[Fe(CN)_6]$ down to the lowest concentrations studied. These ϕ_V^0 values are therefore comparable to ours and agree well, i.e., ϕ_V^0 ($K_4[Mo(CN)_8]$, 25 °C) = 164.5 cm^3 mol^{-1}, ϕ_V^0 ($K_3[Fe(CN)_6]$, 25 °C) = 146.4 cm^3 mol^{-1}. Indelli and co-workers (*1*, *8*) have not analyzed experimental errors; however, they should be similar to ours. An older value[4] of ϕ_V^0 ($K_3[Fe(CN)_6$, 25 °C) = 147.8 cm^3 mol^{-1} appears slightly high.

Billi et al. (*1*) and Hepler et al. (*6*) find that ϕ_V for $K_4[Fe(CN)_6]$ falls sharply to lower values below 10^{-3} mol dm^{-3}. Hepler et al. (*15*) have attributed this behavior to ion-pairing, while Billi et al. (*1*) believe that the Debye-Hückel limiting law may not be applicable to electrolytes containing multiply charged ions (*1*, *7*, *8*) and explain the behavior in terms of the Mayer theory (*7*, *8*). While studying ϕ_V for $K_2[Zn(CN)_4]$ and $K_2[Cd(CN)_4]$ we have observed (*10*) an even more dramatic fall off of ϕ_V values at low concentrations. We have quantitatively accounted for this in terms of ionization and hydrolysis equilibria which become important at high dilutions. We believe similar effects to be the cause of the observed low concentration behavior of $K_4[Fe(CN)_6]$. The sixth stepwise stability constant (*13*) for $K_4[Fe(CN)_6]$ is ca. 10^6 indicating a 10% degree of dissociation at 10^{-4} mol dm^{-3}. Extrapolation to ϕ_V^0 from concentrations below ca. 10^{-3} mol dm^{-3} thus appears to be an unreliable procedure and we prefer the use of the Debye-Hückel limiting law from concentrations $10^{-3} < c < 10^{-2}$ mol dm^{-3}. It should be noted that our values of ϕ_V for $K_4[Fe(CN)_6]$ at concentrations above 10^{-3} mol dm^{-3} are in close agreement with those found by the other workers (*1*, *6*), see Figure 1.

Glossary

c	molar concentration of solution, mol dm^{-3}
d	density of solution, g cm^{-3}
d_0	density of solvent, g cm^{-3}
Δh(corr)	height change in capillary during dilatometer experiment corrected for change in hydrostatic head, cm
$\bar{M}_2$	molecular weight of solute, g mol^{-1}
n_2	amount of solute involved in dilatometer experiment, mol
$\bar{V}_2^0$	partial molar volume of solute at infinite dilution, cm^3 mol^{-1}
Δv	volume change during dilatometer experiment, cm^3

Greek Letters

δ	uncertainty in a quantity, differential
ϕ_V	apparent molar volume, cm^3 mol^{-1}
ϕ_V^0	apparent molar volume at infinite dilution, cm^3 mol^{-1}
ϕ_V^{final}, ϕ_V^{init}	apparent molar volumes of final and initial solutions involved in dilatometer experiment, cm^3 mol^{-1}

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