JOURNAL OF CHEMICAL & ENGINEERING DATA

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Viscosities of the Ternary Systems $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$, $Y(NO_3)_3 + Nd(NO_3)_3 + H_2O$, and $Ce(NO_3)_3 + Nd(NO_3)_3 + H_2O$ and Their Binary Subsystems at Different Temperatures and Atmospheric Pressure

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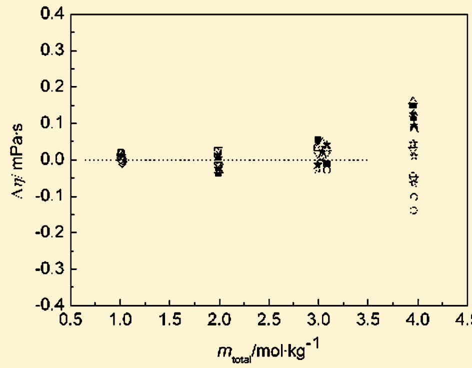
ABSTRACT: Viscosities were measured for the ternary systems $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$, $Y(NO_3)_3 + Nd(NO_3)_3 + H_2O$, and $Ce(NO_3)_3 + Nd(NO_3)_3 + H_2O$ and their binary subsystems $Y(NO_3)_3 + H_2O$, $Ce(NO_3)_3 + H_2O$, and $Nd(NO_3)_3 + H_2O$ at (293.15, 298.15, and 308.15) K and atmospheric pressure. The results were used to test the applicability of simple equations for the viscosity of mixed solutions. The predictions agree well with measured values, implying that the viscosities of the examined electrolyte solutions can be related to those of their constituent binary solutions using these simple equations.



■ INTRODUCTION

The thermodynamic and transport properties of concentrated electrolyte solutions are required for the research and the reliable design of many processes including absorption refrigeration and heat pump systems, distillation with salts, extraction separation, and liquid−liquid dispersions. Up to now extensive data for the thermodynamic and transport properties of binary aqueous solutions have been reported, and recently the viscosities of 74 binary solutions at different temperatures have been tabulated and critically evaluated by Laliberté.[1] However, relatively few measurements have been made for mixed electrolyte solutions. Because the electrolyte solutions encountered in industrial processes appeared to be often the concentrated mixed electrolyte solutions,[1,2] it becomes interesting and practically important to develop simple predictive equations, which can make full use of the available information on the binary electrolyte solutions and provide sufficiently accurate predictions for the mixed electrolyte solutions.

The simple predictive equations have been established for the thermodynamic and transport properties of multicomponent solutions and have been systematically checked by comparisons with the experimental results.[1−15] Recently, excellent reviews of existing viscosity mixing rules and models have been given by Laliberté[1] and Yang et al.[2] In addition, Laliberté[1] proposed a mixing rule and a viscosity model that are applicable to solutions of an arbitrary number of solutes in water, with no limits on solute concentration or solution temperature. The agreements between the predictions of Laliberté's model[1] and the experimental viscosities are very impressive.

Up to now, relatively few measurements have been made for the viscosities of rare earth nitrate solutions, for example, the viscosities of aqueous solutions of $M_1(NO)_3 + M_2(NO)_3$ electrolyte mixtures, where M_i^{3+} (i = 1 and 2) denotes rare earth cations. Therefore, in this study the viscosities were measured for the ternary systems $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$, $Y(NO_3)_3 + Nd(NO_3)_3 + H_2O$, and $Ce(NO_3)_3 + Nd(NO_3)_3 + H_2O$ and their binary subsystems $Y(NO_3)_3 + H_2O$, $Ce(NO_3)_3 + H_2O$, and $Nd(NO_3)_3 + H_2O$ at different temperatures and atmospheric pressure, with $I_{max} \leq 24.5$ mol·kg^{-1} (I is ionic strength). The results thus obtained allow us to extend the tests of the simple viscosity equations *to higher ionic strengths*.

■ EXPERIMENTAL SECTION

Materials. Double-distilled deionized water was prepared by redistillation of distilled water from an alkaline potassium permanganate solution, and its conductivity was (0.8 to 1.2)·10^{-4} S·cm^{-1}.[16] $Y(NO_3)_3·6H_2O$ (with a mass fraction purity of 0.9999), $Ce(NO_3)_3·6H_2O$ (with a mass fraction purity of 0.9999), and $Nd(NO_3)_3·xH_2O$ (with a mass fraction purity of >0.99) were supplied by Shanghai Aladdin Reagent Co., Ltd. The binary solutions were prepared by the method of Spedding et al.[17] Therefore, each rare earth nitrate was dissolved into double-distilled deionized water to prepare the stock solution.[12,18] The resulting solutions were adjusted to their equivalent concentrations with dilute HNO_3 solutions and then reheated and readjusted until stabilized.[12,17,18] The molalities of

Received: August 18, 2011
Accepted: December 22, 2011
Published: January 18, 2012

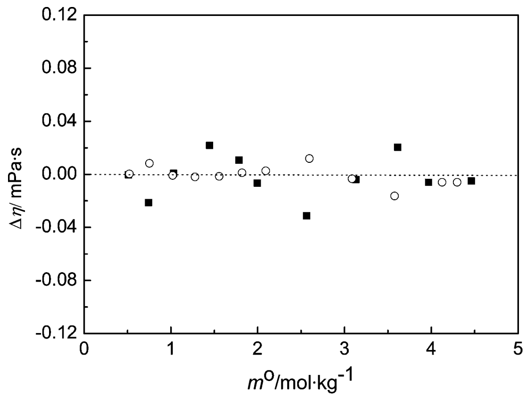


Figure 1. The difference between the present and the literature viscosities ($\Delta\eta = \eta^{o}_{present} - \eta^{o}_{literature}$) as a function of the molality of the rare earth nitrate in the binary solutions at 298.15 K and atmospheric pressure. ■, $\eta^{o}_{present} - \eta^{o}_{ref28}$ ($Ce(NO_3)_3 + H_2O$); ○, $\eta^{o}_{present} - \eta^{o}_{ref29}$ ($Nd(NO_3)_3 + H_2O$).

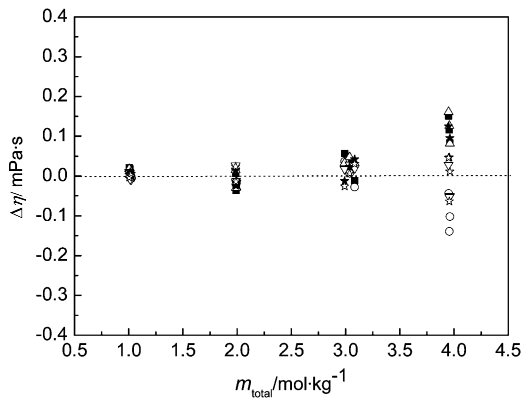


Figure 2. The difference between experimental and predicted viscosities ($\Delta\eta_{eqi} = \eta_{pred.eqi} - \eta_{exp.}$) as a function of the total molality ($m_{total} = m_{Y(NO_3)_3} + m_{Ce(NO_3)_3}$) of the rare earth nitrates in the ternary system $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$ at (293.15, 298.15, and 308.15) K and atmospheric pressure. ■, △, and ★, the values of $\Delta\eta_{eq2}$ at (293.15, 298.15, and 308.15) K; ○, ▽, and ☆, the values of $\Delta\eta_{eq3}$ at (293.15, 298.15, and 308.15) K.

rare earth nitrate stock solutions were analyzed by both ethylenediaminetetraacetic acid (EDTA)[12,18,19] and sulfate methods.[12,17,18] The stock solution concentrations were determined with an accuracy of ≤0.10 %.[12,18,19] Secondary stock solutions of low and moderate concentrations were prepared from weighed quantities of a primary stock solution and double-distilled deionized water using a Sartorius CT225D balance with the precision of $\pm\ 5\cdot10^{-5}$ g.[12,16,18] The ternary solutions were prepared by mixing the binary solutions (the mass of each binary solution is more than 20 g).[20] The uncertainties in the compositions of the prepared solutions due to errors in the weights of binary solutions were estimated to be less than $\pm\ 5\cdot10^{-5}$ mol·kg^{-1}. All solutions were immediately prepared before use, and all of the measurements were performed at atmospheric pressure.

Apparatus and Procedure. Viscosities of solutions were measured using modified Cannon–Ubbelohde suspended level capillary viscometers of (0.4, 0.6, 0.8, and 0.8) mm in diameters. The viscometer constant is effectively independent of temperature.[20,21] No surface tension corrections are required.[20,21] The tops of these viscometers were modified slightly to allow the interior of the viscometer to be sealed off from the atmosphere, yet allowing the suspended-level feature to be retained. A thoroughly cleaned and perfectly dried viscometer filled with liquid was placed vertically in a glass sided water thermostat. The temperature in the water thermostat was kept constant and monitored by a DP95 digital RTD thermometer (ITS-90) with an uncertainty of ± 0.01 K.[16,20] After thermal equilibrium was attained the efflux time of flow of the liquids were recorded with a digital stop watch with a precision of ± 0.01 s.[16,20] The capillary viscometers were calibrated and credited by the company (the values of the viscometer constant are (0.003636, 0.01183, 0.03023, and 0.0511) mm^2·s^{-2}, respectively),[16,20] and with a stated precision of ± 0.1 %.[16,20] The viscosity of the solution is given by[14,20,22]

$$\eta = \eta_o \frac{\rho\tau}{\rho_o\tau_o} \quad (1)$$

Table 1. Experimental Viscosities of the Binary Solutions $Y(NO_3)_3 + H_2O$, $Ce(NO_3)_3 + H_2O$, and $Nd(NO_3)_3 + H_2O$ at Different Temperatures and Atmospheric Pressure

m	I	$\eta^{o}_{293.15}$	$\eta^{o}_{298.15}$/mPa·s		$\eta^{o}_{303.15}$
mol·kg^{-1}	mol·kg^{-1}	mPa·s	exptl	ref	mPa·s
		$Y(NO_3)_3 + H_2O$			
0.4989	2.9934	1.334_9	1.190_7		0.965_5
0.7348	4.4088	1.556_3	1.381_9		1.134_7
0.9965	5.9790	1.809_3	1.609_6		1.304_8
1.2803	7.6818	2.149_7	1.913_9		1.537_0
1.4972	8.9832	2.473_6	2.197_3		1.746_7
1.7235	10.3410	2.892_9	2.555_9		2.023_6
1.9820	11.8920	3.483_9	3.056_2		2.404_5
2.5432	15.2592	5.299_1	4.585_8		3.559_1
2.9475	17.6850	7.180_1	6.163_9		4.676_5
3.4569	20.7414	10.527	8.957_2		6.536_2
3.9452	23.6712	15.240	12.798		8.990_7
4.9575	29.7450	33.505	26.724		18.139
		$Ce(NO_3)_3 + H_2O$			
0.5112	3.0672	1.311_7	1.173_6	1.173_9[28]	0.950_9
0.7425	4.4550	1.441_9	1.278_8	1.300_1[28]	1.053_5
1.0313	6.1878	1.717_7	1.529_2	1.528_5[28]	1.248_8
1.4465	8.6790	2.275_5	2.037_1	2.015_3[28]	1.629_6
1.7852	10.7112	2.848_3	2.545_4	2.534_7[28]	2.017_4
1.9969	11.9814	3.271_6	2.901_9	2.908_5[28]	2.298_9
2.5634	15.3804	4.702_3	4.098_8	4.130_1[28]	3.228_9
3.1308	18.7848	6.898_5	5.937_5	5.941_4[28]	4.599_3
3.6134	21.6804	9.857_1	8.409_1	8.388_7[28]	6.328_6
3.9676	23.8056	13.018	10.989	10.995[28]	8.054_8
4.4616	26.7696	19.480	16.059	16.064[28]	11.338
		$Nd(NO_3)_3 + H_2O$			
0.5220	3.1320	1.299_0	1.159_0	1.158_7[29]	0.943_3
0.7543	4.5258	1.485_9	1.323_3	1.315_1[29]	1.076_0
1.0213	6.1278	1.721_2	1.529_8	1.530_6[29]	1.241_6
1.2786	7.6716	1.997_6	1.779_0	1.781_0[29]	1.439_1
1.5554	9.3324	2.369_5	2.105_7	2.107_3[29]	1.696_2
1.8203	10.9218	2.815_6	2.488_2	2.487_1[29]	1.983_2
2.0934	12.5604	3.382_5	2.966_5	2.963_9[29]	2.352_6
2.5973	15.5838	4.798_7	4.154_5	4.142_6[29]	3.227_2
3.0857	18.5142	6.774_5	5.799_7	5.803_1[29]	4.380_2
3.5762	21.4572	9.719_9	8.216_3	8.232_7[29]	6.019_7
4.1242	24.7452	14.901	12.341	12.347[29]	8.714_7
4.2972	25.7832	17.157	14.090	14.096[29]	9.848_4

Table 2. Parameters for the Binary Solutions $Y(NO_3)_3$ + H_2O, $Ce(NO_3)_3$ + H_2O, and $Nd(NO_3)_3$ + H_2O at Different Temperatures and Atmospheric Pressure

T/K	293.15	298.15	308.15		293.15	298.15	308.15
		$\eta^o_{Y(NO_3)_3+H_2O}$				$\eta^o_{Ce(NO_3)_3+H_2O}$	
A_0	0.689867	0.730354	0.389535	A_0	1.40058	1.47514	0.99791
	±0.009212	±0.009913	±0.023012		±0.014573	±0.017174	±0.016853
A_1	1.80105	1.13907	1.70936	A_1	−0.811723	−1.5756	−0.570334
	±0.028229	±0.030377	±0.07052		±0.045467	±0.053582	±0.052580
A_2	−1.45149	−0.649349	−1.52409	A_2	1.431310	2.36858	1.12472
	±0.029423	±0.031662	±0.07350		±0.048659	±0.057345	±0.056272
A_3	0.969488	0.476969	0.922366	A_3	−0.384983	−0.947845	−0.405122
	±0.013477	±0.014503	±0.033666		±0.023190	±0.027330	±0.026818
A_4	−0.218382	−0.0939361	−0.204282	$10A_4$	0.405924	1.8461	0.799777
	±0.002784	±0.002996	±0.006955		±0.050399	±0.059395	±0.058284
$10A_5$	0.244936	0.116654	0.192833	10^2A_5	0.640088	−0.820508	−0.29498
	±0.002112	±0.002273	±0.005276		±0.040646	±0.047901	±0.047004
δ^o_η	$2.8\cdot10^{-4}$	$3.4\cdot10^{-4}$	$9.9\cdot10^{-4}$	δ^o_η	$3.8\cdot10^{-4}$	$4.4\cdot10^{-4}$	$5.3\cdot10^{-3}$
SSE[a]	$1.2\cdot10^{-5}$	$1.4\cdot10^{-5}$	$7.6\cdot10^{-5}$	SSE[a]	$1.9\cdot10^{-5}$	$2.6\cdot10^{-5}$	$2.5\cdot10^{-5}$
RMSE[a]	$1.4\cdot10^{-3}$	$1.5\cdot10^{-3}$	$3.6\cdot10^{-3}$	RMSE[a]	$1.9\cdot10^{-3}$	$2.3\cdot10^{-3}$	$2.3\cdot10^{-3}$
		$\eta^o_{Nd(NO_3)_3+H_2O}$			$\varphi^{o,298.15}_{Y(NO_3)_3+H_2O}$	$\varphi^{o,298.15}_{Ce(NO_3)_3+H_2O}$	$\varphi^{o,298.15}_{Nd(NO_3)_3+H_2O}$
A_0	0.728285	0.766456	0.611172	B_0	0.816929	−3.51640	0.760181
	±0.022306	±0.019880	±0.032721		±0.009203	±0.53181	±0.005866
A_1	1.53507	0.918193	0.792068	B_1	−0.343762	15.4662	−0.131740
	±0.070819	±0.063114	±0.103885		±0.043732	±1.8434	±0.029410
A_2	−1.26525	−0.526753	−0.477658	B_2	0.722643	−21.8132	0.132389
	±0.078294	±0.069776	±0.114851		±0.073484	±2.4977	±0.052327
A_3	0.907545	0.456174	0.389048	B_3	−0.164927	15.0855	0.207792
	±0.038774	±0.034555	±0.056878		±0.055676	±1.6555	±0.042137
$10A_4$	−2.30653	−1.14354	−0.961252	$10B_4$	0.10871	−49.9990	−1.02828
	±0.087846	±0.078289	±0.128863		±0.194183	±5.3731	±0.156646
$10A_5$	0.271817	0.149452	0.113003	10^3B_5	−0.546332	643.077	14.0003
	±0.007400	±0.006595	±0.010855		±2.53488	±68.407	±2.18397
δ^o_η	$5.5\cdot10^{-4}$	$6.0\cdot10^{-4}$	$9.8\cdot10^{-4}$	δ^o_φ	$9.7\cdot10^{-4}$	$4.5\cdot10^{-3}$	$6.5\cdot10^{-4}$
SSE[a]	$4.6\cdot10^{-5}$	$3.6\cdot10^{-5}$	$9.9\cdot10^{-5}$	SSE[a]	$9.8\cdot10^{-5}$	$8.5\cdot10^{-6}$	$2.5\cdot10^{-5}$
RMSE[a]	$2.8\cdot10^{-3}$	$2.5\cdot10^{-3}$	$4.1\cdot10^{-3}$	RMSE[a]	$1.6\cdot10^{-3}$	$1.3\cdot10^{-3}$	$9.0\cdot10^{-4}$

[a] Goodness of fit statistics: SSE, the sum of squares due to error; RMSE, the root-mean-squared error; R-squared, the coefficient of multiple determination (not less than 0.9999 in this work); adjusted R-squared, the degree of freedom adjusted R-squared (not less than 0.9999 in this work). The confidence bounds for fitted coefficients are 95 %.

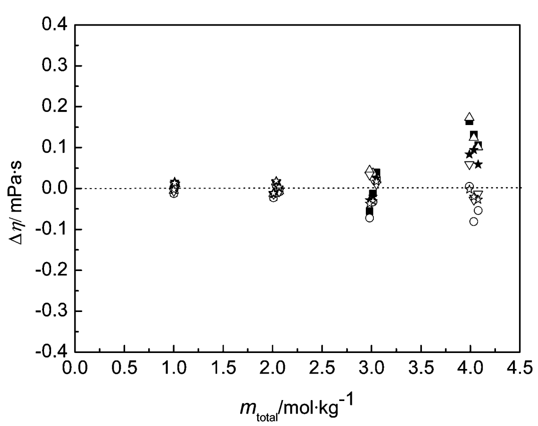


Figure 3. The difference between experimental and predicted viscosities ($\Delta\eta_{eqi} = \eta_{pred.eqi} - \eta_{exp.}$) as a function of the total molality ($m_{total} = m_{Y(NO_3)_3} + m_{Nd(NO_3)_3}$) of the rare earth nitrates in the ternary system $Y(NO_3)_3$ + $Nd(NO_3)_3$ + H_2O at (293.15, 298.15, and 308.15) K and atmospheric pressure. ■, △, and ★, the values of $\Delta\eta_{eq2}$ at (293.15, 298.15, and 308.15) K; ○, ▽, and ☆, the values of $\Delta\eta_{eq3}$ at (293.15, 298.15, and 308.15) K.

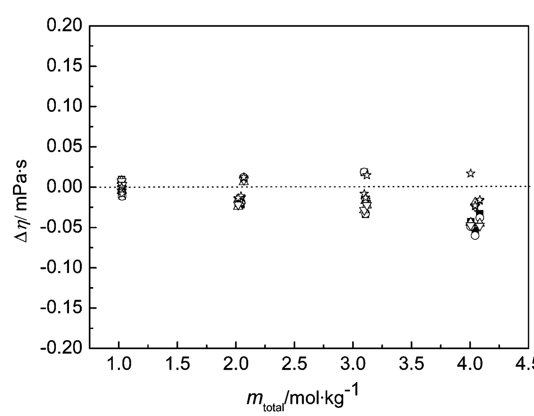


Figure 4. The difference between experimental and predicted viscosities ($\Delta\eta_{eqi} = \eta_{pred.eqi} - \eta_{exp.}$) as a function of the total molality ($m_{total} = m_{Ce(NO_3)_3} + m_{Nd(NO_3)_3}$) of the rare earth nitrates in the ternary system $Ce(NO_3)_3$ + $Nd(NO_3)_3$ + H_2O at (293.15, 298.15, and 308.15) K and atmospheric pressure. ■, △, and ★, the values of $\Delta\eta_{eq2}$ at (293.15, 298.15, and 308.15) K; ○, ▽, and ☆, the values of $\Delta\eta_{eq3}$ at (293.15, 298.15, and 308.15) K.

dx.doi.org/10.1021/je200914j | J. Chem. Eng.Data 2012, 57, 366−373

Table 3. Comparisons of Measured and Predicted Viscosities of the Ternary System $Y(NO_3)_3$ (B) + $Ce(NO_3)_3$ (C) + H_2O at Different Temperatures and Atmospheric Pressure, with $I_{max} \leq 23.8$ mol·kg^{-1}

			η			$\Delta\eta$	
m_B	m_C	I	mPa·s			mPa·s	
mol·kg^{-1}	mol·kg^{-1}	mol·kg^{-1}	exp	eq 2	eq 3	Δ_{eq} 2	Δ_{eq} 3
			293.15 K				
0.2545	0.7679	6.1344	1.745_8	1.746_4	1.738_8	0.000_6	-0.007_0
0.5027	0.5111	6.0828	1.753_9	1.771_8	1.761_7	0.017_9	0.007_8
0.7527	0.2523	6.0300	1.773_8	1.793_7	1.786_1	0.019_9	0.012_3
0.4944	1.4988	11.9592	3.340_8	3.312_9	3.323_2	-0.027_9	-0.017_6
0.9897	0.9998	11.9370	3.397_6	3.361_7	3.376_7	-0.035_9	-0.020_9
1.4843	0.5014	11.9142	3.413_0	3.418_5	3.430_3	0.005_5	0.017_3
0.7655	2.3177	18.4992	6.996_5	6.985_1	6.968_7	-0.011_4	-0.027_8
1.4879	1.5503	18.2292	7.030_3	7.061_0	7.036_7	0.030_7	0.006_4
2.2402	0.7513	17.9490	7.073_8	7.130_4	7.110_6	0.056_6	0.036_8
0.9855	2.9766	23.7726	13.640	13.723	13.538	0.083	−0.102
1.9734	1.9830	23.7384	14.219	14.335	14.080	0.116	−0.139
2.9596	0.9912	23.7048	14.691	14.842	14.647	0.151	−0.044
					$\delta_\eta^{eqi\,a}$	$6.7\cdot10^{-3}$	$5.2\cdot10^{-3}$
			298.15 K				
0.2545	0.7679	6.1344	1.556_6	1.555_5	1.547_9	-0.001_1	-0.008_7
0.5027	0.5111	6.0828	1.575_8	1.578_0	1.567_8	0.002_2	-0.008_0
0.7527	0.2523	6.0300	1.579_2	1.596_8	1.589_1	0.017_6	0.009_9
0.4944	1.4988	11.9592	2.957_3	2.928_9	2.940_1	-0.028_4	-0.017_2
0.9897	0.9998	11.9370	2.992_8	2.963_4	2.979_2	-0.029_4	-0.013_6
1.4843	0.5014	11.9142	2.991_6	3.005_9	3.018_3	0.014_3	0.026_7
0.7655	2.3177	18.4992	5.976_7	6.008_6	5.996_1	0.031_9	0.019_4
1.4879	1.5503	18.2292	6.022_2	6.069_5	6.050_9	0.047_3	0.028_7
2.2402	0.7513	17.9490	6.092_7	6.125_2	6.110_1	0.032_5	0.017_4
0.9855	2.9766	23.7726	11.464	11.546	11.413	0.082	−0.051
1.9734	1.9830	23.7384	11.908	12.034	11.855	0.126	−0.053
2.9596	0.9912	23.7048	12.289	12.450	12.317	0.161	0.028
					$\delta_\eta^{eqi\,a}$	$7.2\cdot10^{-3}$	$4.9\cdot10^{-3}$
			308.15 K				
0.2545	0.7679	6.1344	1.263_4	1.268_4	1.262_6	0.005_0	-0.000_8
0.5027	0.5111	6.0828	1.274_7	1.285_1	1.277_3	0.010_4	0.002_6
0.7527	0.2523	6.0300	1.296_9	1.299_0	1.293_1	0.002_1	-0.003_8
0.4944	1.4988	11.9592	2.333_8	2.316_6	2.324_2	-0.017_2	-0.009_6
0.9897	0.9998	11.9370	2.366_3	2.341_6	2.352_5	-0.024_7	-0.013_8
1.4843	0.5014	11.9142	2.357_0	2.372_1	2.380_6	0.015_1	0.023_6
0.7655	2.3177	18.4992	4.586_9	4.629_0	4.617_2	0.042_1	0.030_3
1.4879	1.5503	18.2292	4.628_8	4.652_0	4.635_0	0.023_2	0.006_2
2.2402	0.7513	17.9490	4.682_6	4.669_8	4.656_3	-0.012_8	-0.026_3
0.9855	2.9766	23.7726	8.267_5	8.362_5	8.278_6	0.095_0	0.011_1
1.9734	1.9830	23.7384	8.571_9	8.618_5	8.508_1	0.046_6	-0.063_8
2.9596	0.9912	23.7048	8.700_1	8.825_1	8.745_6	0.125_0	0.045_5
					$\delta_\eta^{eqi\,a}$	$7.2\cdot10^{-3}$	$4.4\cdot10^{-3}$

$^a\delta_\eta^{eqi} = \sum_{j=1}^{N}(|\eta_{j(eqi)} - \eta_{j(exp)}|/\eta_{j(exp)})/N$.

where η_o is the viscosity of water. ρ and ρ_o are the densities of the solution and water, respectively. τ and τ_o are the flow time of the solution and water, respectively. The densities of the solutions used in this research were taken from ref 23. Triplicate measurements were performed at each composition. The average value is reported. As a check on the accuracy of the method used, the viscosity of water at 298.15 K was determined and compared with the literature value (the density of water at this temperature was taken as 0.99701 g·cm^{-1}[24,25]). The result is 0.8905 versus 0.8903 mPa·s.[24−27] The uncertainty in viscosity is $\pm$ $2\cdot10^{-4}$ mPa·s. Further checks on the accuracy of the present method were made using the literature viscosities of the binary solutions $Ce(NO_3)_3$ + H_2O and $Nd(NO_3)_3$ + H_2O. The comparisons are shown in Table 1. The uncertainty in the temperature, including fluctuations and temperature gradients, is estimated to be 0.02 K. This leads to a relative standard uncertainty of 0.1 % in the viscosity measurement. The uncertainty in the flow time measurement is estimated to be less than 0.1 s, which leads to a relative standard uncertainty of less than 0.03 % in the viscosity measurement. The uncertainty in the calibration constant for the viscometer leads to a relative standard uncertainty of less than 0.5 % in the viscosity measurement. The accuracy of the density measurements was $\pm$ $5\cdot10^{-5}$ g·cm^{-3},[23] which leads to a relative standard uncertainty of less than 0.09 % in the viscosity measurement. After using standard techniques for the propagation of

dx.doi.org/10.1021/je200914j | J. Chem. Eng.Data 2012, 57, 366−373

Table 4. Comparisons of Measured and Predicted Viscosities of the Ternary System $Y(NO_3)_3$ (B) + $Nd(NO_3)_3$ (C) + H_2O at Different Temperatures and Atmospheric Pressure, with $I_{max} \leq 24.5$ mol·kg^{-1}

			η			$\Delta\eta$	
m_B	m_C	I	mPa·s			mPa·s	
mol·kg^{-1}	mol·kg^{-1}	mol·kg^{-1}	exp	eq 2	eq 3	Δ_{eq} 2	Δ_{eq} 3
			293.15 K				
0.2532	0.7618	6.0900	1.735_9	1.746_1	1.744_3	0.010_2	0.008_4
0.5005	0.5083	6.0528	1.759_6	1.768_5	1.766_1	0.008_9	0.006_5
0.7512	0.2514	6.0156	1.801_2	1.790_6	1.788_7	-0.010_6	-0.012_5
0.5132	1.5514	12.3876	3.420_5	3.412_9	3.411_6	-0.007_6	-0.008_9
1.0147	1.0217	12.2184	3.428_3	3.438_2	3.436_6	0.009_9	0.008_3
1.5034	0.5055	12.0534	3.483_7	3.462_1	3.461_0	-0.021_6	-0.022_7
0.7595	2.2906	18.3006	6.849_7	6.888_9	6.873_9	0.039_2	0.024_2
1.5011	1.5142	18.0918	7.007_1	6.995_1	6.974_0	-0.012_0	-0.033_1
2.2355	0.7454	17.8854	7.149_2	7.093_7	7.077_2	-0.055_5	-0.072_0
1.0125	3.0658	24.4698	15.005	15.111	14.951	0.106	−0.054
2.0077	2.0254	24.1986	15.104	15.236	15.023	0.132	−0.081
2.9764	1.0127	23.9346	15.114	15.278	15.119	0.164	0.005
					$\delta_\eta^{eqi\,a}$	$5.8\cdot10^{-3}$	$4.6\cdot10^{-3}$
			298.15 K				
0.2532	0.7618	6.0900	1.542_9	1.553_8	1.552_2	0.010_9	0.009_3
0.5005	0.5083	6.0528	1.561_9	1.573_6	1.571_5	0.011_7	0.009_6
0.7512	0.2514	6.0156	1.598_0	1.593_1	1.591_5	-0.004_9	-0.006_5
0.5132	1.5514	12.3876	3.000_0	2.993_6	2.992_6	-0.006_4	-0.007_4
1.0147	1.0217	12.2184	3.000_4	3.016_1	3.014_9	0.015_7	0.014_5
1.5034	0.5055	12.0534	3.050_2	3.037_4	3.036_6	-0.012_8	-0.013_6
0.7595	2.2906	18.3006	5.879_6	5.900_1	5.890_3	0.020_5	0.010_7
1.5011	1.5142	18.0918	5.957_4	5.994_6	5.980_8	0.037_2	0.023_4
2.2355	0.7454	17.8854	6.040_1	6.084_2	6.073_4	0.044_1	0.033_3
1.0125	3.0658	24.4698	12.441	12.542	12.428	0.101	−0.013
2.0077	2.0254	24.1986	12.561	12.685	12.533	0.124	−0.028
2.9764	1.0127	23.9346	12.596	12.769	12.655	0.173	0.059
					$\delta_\eta^{eqi\,a}$	$6.5\cdot10^{-3}$	$3.9\cdot10^{-3}$
			308.15 K				
0.2532	0.7618	6.0900	1.257_7	1.262_6	1.260_2	0.004_9	0.002_5
0.5005	0.5083	6.0528	1.263_2	1.279_4	1.276_1	0.016_2	0.012_9
0.7512	0.2514	6.0156	1.295_9	1.295_3	1.292_8	-0.000_6	-0.003_1
0.5132	1.5514	12.3876	2.366_3	2.366_6	2.368_9	0.000_3	0.002_6
1.0147	1.0217	12.2184	2.365_0	2.380_2	2.383_4	0.015_2	0.018_4
1.5034	0.5055	12.0534	2.408_3	2.394_3	2.396_7	-0.014_0	-0.011_6
0.7595	2.2906	18.3006	4.438_6	4.463_1	4.455_5	0.024_5	0.016_9
1.5011	1.5142	18.0918	4.559_1	4.539_1	4.528_0	-0.020_0	-0.031_1
2.2355	0.7454	17.8854	4.640_0	4.611_7	4.602_7	-0.028_3	-0.037_3
1.0125	3.0658	24.4698	8.794_3	8.852_8	8.766_6	0.058_5	-0.027_7
2.0077	2.0254	24.1986	8.849_1	8.943_0	8.828_4	0.093_9	-0.020_7
2.9764	1.0127	23.9346	8.904_7	8.988_2	8.902_4	0.083_5	-0.002_3
					$\delta_\eta^{eqi\,a}$	$6.0\cdot10^{-3}$	$4.4\cdot10^{-3}$

[a] $\delta_\eta^{eqi} = \sum_{j=1}^{N}(|\eta_{j(eqi)} - \eta_{j(exp)}|/\eta_{j(exp)})/N$.

uncertainty, the estimated overall expanded uncertainty, including efflux time, temperature, the accuracy of the density measurement, and calibration uncertainties, is 1.0 % ($k = 2$).

■ RESULTS AND DISCUSSION

Simple Predictive Equations for Viscosity. In the following section, the variables with the superscript o together with the subscript $M_i(NO_3)_3$ were used to denote the quantities of component $M_i(NO_3)_3$ in the binary solution $M_i(NO_3)_3$ + H_2O (i = 1 and 2) having the same water activity as that of a mixed solution, and those without the superscript (io) denote the corresponding quantities in the mixed solution.

According to the semi-ideal solution theory,[3−6,15] the viscosity of a ternary solution is related to those of its constituent binary solutions of equal water activity by

$$\ln \eta = \sum_i (x_{M_i(NO_3)_3}/x^{(io)}_{M_i(NO_3)_3})\ln \eta^{(io)}_{M_i(NO_3)_3} \tag{2}$$

where $x^{(io)}_{M_i(NO_3)_3}$ and $\eta^{(io)}_{M_i(NO_3)_3}$ denote the mole fraction and the viscosity of the binary solution $M_i(NO_3)_3$ + H_2O (i = 1 and 2) having the same water activity as that of the mixed solution $M_1(NO_3)_3$ + $M_2(NO_3)_3$ + H_2O. $x_{M_i(NO_3)_3}$ is the mole fraction of $M_i(NO_3)_3$ in the mixed solution.

Table 5. Comparisons of Measured and Predicted Viscosities of the Ternary System $Ce(NO_3)_3$ (B) + $Nd(NO_3)_3$ (C) + H_2O at Different Temperatures and Atmospheric Pressure, with $I_{max} \leq 24.5$ mol·kg^{-1}

m_B	m_C	I	η			$\Delta\eta$	
			mPa·s			mPa·s	
mol·kg^{-1}	mol·kg^{-1}	mol·kg^{-1}	exp	eq 2	eq 3	Δ_{eq} 2	Δ_{eq} 3
			293.15 K				
0.2565	0.7673	6.1428	1.711_9	1.721_1	1.720_7	0.009_2	0.008_8
0.5097	0.5165	6.1572	1.725_0	1.719_7	1.719_1	-0.005_3	-0.005_9
0.7720	0.2568	6.1728	1.729_2	1.718_3	1.717_9	-0.010_9	-0.011_3
0.5182	1.5501	12.4098	3.339_8	3.349_4	3.352_0	0.009_6	0.012_2
1.0233	1.0207	12.2640	3.340_3	3.318_6	3.321_9	-0.021_7	-0.018_4
1.5159	0.5042	12.1206	3.312_8	3.292_1	3.294_5	-0.020_7	-0.018_3
0.7723	2.3246	18.5814	6.787_8	6.806_1	6.806_6	0.018_3	0.018_8
1.5687	1.5396	18.6498	6.870_9	6.837_3	6.838_0	-0.033_6	-0.032_9
2.3415	0.7779	18.7164	6.888_5	6.868_0	6.868_5	-0.020_5	-0.020_0
1.0189	3.0651	24.5040	14.435	14.402	14.397	−0.033	−0.038
2.0174	2.0272	24.2676	13.977	13.923	13.917	−0.054	−0.06
3.0001	1.0057	24.0348	13.506	13.463	13.457	−0.043	−0.049
					$\delta_\eta^{eqi\,a}$	$4.2 \cdot 10^{-3}$	$4.2 \cdot 10^{-3}$
			298.15 K				
0.2565	0.7673	6.1428	1.524_4	1.532_0	1.531_6	0.007_6	0.007_2
0.5097	0.5165	6.1572	1.535_7	1.531_0	1.530_4	-0.004_7	-0.005_3
0.7720	0.2568	6.1728	1.526_7	1.530_1	1.529_6	0.003_4	0.002_9
0.5182	1.5501	12.4098	2.939_4	2.945_6	2.948_2	0.006_2	0.008_8
1.0233	1.0207	12.2640	2.943_7	2.926_6	2.930_0	-0.017_1	-0.013_7
1.5159	0.5042	12.1206	2.935_9	2.911_8	2.914_2	-0.024_1	-0.021_7
0.7723	2.3246	18.5814	5.863_7	5.835_1	5.835_2	-0.028_6	-0.028_5
1.5687	1.5396	18.6498	5.887_2	5.870_3	5.870_4	-0.016_9	-0.016_8
2.3415	0.7779	18.7164	5.927_2	5.904_7	5.904_8	-0.022_5	-0.022_4
1.0189	3.0651	24.5040	12.031	11.986	11.983	−0.045	−0.048
2.0174	2.0272	24.2676	11.663	11.644	11.639	−0.019	−0.024
3.0001	1.0057	24.0348	11.356	11.312	11.308	−0.044	−0.048
					$\delta_\eta^{eqi\,a}$	$3.9 \cdot 10^{-3}$	$3.9 \cdot 10^{-3}$
			308.15 K				
0.2565	0.7673	6.1428	1.245_5	1.245_4	1.245_2	-0.000_1	-0.000_3
0.5097	0.5165	6.1572	1.251_0	1.246_4	1.246_1	-0.004_6	-0.004_9
0.7720	0.2568	6.1728	1.255_7	1.247_6	1.247_3	-0.008_1	−0.0084
0.5182	1.5501	12.4098	2.324_5	2.335_6	2.336_8	0.011_1	0.012_3
1.0233	1.0207	12.2640	2.333_2	2.320_5	2.322_0	-0.012_7	-0.011_2
1.5159	0.5042	12.1206	2.321_9	2.307_2	2.308_3	-0.014_7	-0.013_6
0.7723	2.3246	18.5814	4.445_3	4.437_2	4.437_0	-0.008_1	-0.008_3
1.5687	1.5396	18.6498	4.504_8	4.491_9	4.491_6	-0.012_9	-0.013_2
2.3415	0.7779	18.7164	4.530_6	4.545_6	4.545_3	0.015_0	0.014_7
1.0189	3.0651	24.5040	8.555_5	8.538_6	8.539_7	-0.016_9	-0.015_8
2.0174	2.0272	24.2676	8.394_0	8.370_3	8.371_1	-0.023_7	-0.022_9
3.0001	1.0057	24.0348	8.193_2	8.210_0	8.210_2	0.016_8	0.017_0
					$\delta_\eta^{eqi\,a}$	$3.5 \cdot 10^{-3}$	$3.5 \cdot 10^{-3}$

$^a\delta_\eta^{eqi} = \sum_{j=1}^{N}(|\eta_{j(eqi)} - \eta_{j(exp)}|/\eta_{j(exp)})/N$.

Hu derived the following equation for the viscosity of mixed electrolyte solutions:[11]

$$\ln \eta = \sum_i (x_{M_i(NO_3)_3}/x^{o,I}_{M_i(NO_3)_3})\ln \eta^{o,I}_{M_i(NO_3)_3} \tag{3}$$

where $x^{o,I}_{M_i(NO_3)_3}$ and $\eta^{o,I}_{M_i(NO_3)_3}$ denote the mole fraction and the viscosity of the binary solution $M_i(NO_3)_3 + H_2O$ (i = 1 and 2) having the same ionic strength as that of the mixed solution.

Comparisons of the Measured Viscosities with the Values Reported in the Literature. Table 1 shows the measured viscosities of the binary solutions $Y(NO_3)_3 + H_2O$, $Ce(NO_3)_3 + H_2O$, and $Nd(NO_3)_3 + H_2O$ at different temperatures and atmospheric pressure. The viscosities at 298.15 K are compared with the values calculated from the viscosity equations reported in the literature[28,29] and the molalities shown in Table 1. It can be seen from Table 1 and Figure 1 that the agreements are good.

Test Procedure. The measured viscosities were used to test eqs 2 and 3, and the test procedure is briefly summarized as follows:

(1) Represent the measured viscosities of the binary solutions by the following polynomial equations using MATLAB 7.0 (function of nonlinear fitting):

$$\eta^{o}_{M_i(NO_3)_3(calc)} = \sum_{l=0}^{N} A_l (m^{o}_{M_i(NO_3)_3})^l \quad (4)$$

where $\eta^{o}_{M_i(NO_3)_3(calc)}$ and $m^{o}_{M_i(NO_3)_3}$ denote the viscosity and molality of the binary aqueous solution $M_i(NO_3)_3 + H_2O$ (i = 1 and 2). The optimum fit was obtained by variation of l until the values of $\delta^{o}_{\eta,M_i(NO_3)_3} = \sum_{j=1}^{N}(|\eta^{o}_{M_i(NO_3)_3(calc)} - \eta^{o}_{M_i(NO_3)_3(exp)}|/\eta^{o}_{M_i(NO_3)_3(exp)})/N$ is less than a few parts in 10^{-3}. The values of A_l and $\delta^{o}_{\eta,M_i(NO_3)_3}$ obtained for the three binary solutions are shown in Table 2.

(2) Represent the reported osmotic coefficient data[30−32] by the equations $\varphi^{o}_{M_i(NO_3)_3(calc)} = \sum_{l=0}^{N} B_l (m^{o}_{M_i(NO_3)_3})^{l/2}$. The obtained values of B_l are shown in Table 2. Then, determine the compositions ($m^{(io)}_{M_i(NO_3)_3}$) of the binary solutions having the same water activity as that of the mixed solution of given molalities $m_{M_i(NO_3)_3}$ (i = 1 and 2) using the osmotic coefficients of $M_i(NO_3)_3$ (i = 1 and 2)[30−32] and the linear isopiestic relation:[3−6,33]

$$\frac{m_{M_1(NO_3)_3}}{m^{(1o)}_{M_1(NO_3)_3}} + \frac{m_{M_2(NO_3)_3}}{m^{(2o)}_{M_2(NO_3)_3}} = 1$$
$$\left(a_w = \text{constant and } 0 \le \frac{m_{M_i(NO_3)_3}}{m^{(io)}_{M_i(NO_3)_3}} \le 1\right) \quad (5)$$

where $m_{M_i(NO_3)_3}$ and $m^{(io)}_{M_i(NO_3)_3}$ are the molalities of $M_i(NO_3)_3$ in the mixed aqueous solution $M_1(NO_3)_3 + M_2(NO_3)_3 + H_2O$ and its binary subsystems $M_i(NO_3)_3 + H_2O$ (i = 1 and 2) of equal water activity. It is notable that the osmotic coefficients of its binary subsystems at 298.15 K are used to calculate the compositions ($m^{o}_{M_i(NO_3)_3}$) of the binary solutions at (293.15 and 308.15) K.

(3) Determine the compositions ($m^{o,I}_{M_i(NO_3)_3}$) of the binary solutions having the same ionic strength as that of the mixed solution of given molalities $m_{M_i(NO_3)_3}$ (i = 1 and 2).

(4) Insert the values of $\eta^{(io)}_{M_i(NO_3)_3}$ and $\eta^{o,I}_{M_i(NO_3)_3}$ calculated from eq 4 into eqs 2 and 3 to yield the predictions for the mixed solutions of given $m_{M_i(NO_3)_3}$ (i = 1 and 2), which are then compared with the corresponding experimental data.

In this paper, the average relative differences between the predicted and measured viscosities (δ_η) over the entire experimental composition range of the mixed solution are defined by

$$\delta_\eta = \sum_{i=1}^{N} |\delta_{\eta,i}|/N \quad (6)$$

with $\delta_{\eta,i} = (\eta_{i,(calc)} - \eta_{i,(exp)})/\eta_{i,(exp)}$, where N is the number of experimental data.

Verifications of Equations. Table 3 and Figure 2 compare the predicted and measured viscosities for the ternary solution $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$ at different temperatures and atmospheric pressure. The δ_η values of eqs 2 and 3 are $6.7\cdot10^{-3}$ versus $5.2\cdot10^{-3}$ at 293.15 K, $7.2\cdot10^{-3}$ versus $4.9\cdot10^{-3}$ at 298.15 K, and $7.2\cdot10^{-3}$ versus $4.4\cdot10^{-3}$ at 308.15 K, respectively.

The predicted and measured viscosities are compared in Table 4 and Figure 3 for the ternary solution $Y(NO_3)_3 + Nd(NO_3)_3 + H_2O$ at different temperatures and atmospheric pressure. The δ_η values of eqs 2 and 3 are $5.8\cdot10^{-3}$ versus $4.6\cdot10^{-3}$ at 293.15 K, $6.5\cdot10^{-3}$ versus $3.9\cdot10^{-3}$ at 298.15 K, and $6.0\cdot10^{-3}$ versus $4.4\cdot10^{-3}$ at 308.15 K, respectively.

As can be seen from Table 5 and Figure 4, the δ_η values of eqs 2 and 3 for the system $Ce(NO_3)_3 + Nd(NO_3)_3 + H_2O$ at the three temperatures and atmospheric pressure are the same, that is, $4.2\cdot10^{-3}$ at 293.15 K, $3.9\cdot10^{-3}$ at 298.15 K, and $3.5\cdot10^{-3}$ at 308.15 K, respectively.

■ CONCLUSIONS

The viscosities for the ternary systems $Y(NO_3)_3 + Ce(NO_3)_3 + H_2O$, $Y(NO_3)_3 + Nd(NO_3)_3 + H_2O$, and $Ce(NO_3)_3 + Nd(NO_3)_3 + H_2O$ and their binary subsystems at (293.15, 298.15, and 308.15) K and atmospheric pressure were measured and used to test the predictability of the simple predictive equations. The comparison results show that these simple equations can provide good predictions for the viscosities of the ternary rare earth nitrate solutions in terms of the properties of their binary solutions. In addition, the osmotic coefficients of the binary solutions of rare earth nitrates at 298.15 K can be used to determine the compositions of the ternary rare earth nitrate solutions of equal water activities at other temperatures.

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Funding

The authors thank the National Natural Science Foundation of China (20976189, 21076224, and 21036008) and Science Foundation of China University of Petroleum, Beijing (qzdx-2011-01) for financial support.

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