Measurement and Modeling of Density, Kinematic Viscosity, and Refractive Index for Poly(ethylene Glycol) Aqueous Solution at Different Temperatures

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The density, kinematic viscosity, and refractive index of aqueous solutions of poly(ethylene glycol) (PEG) have been measured at (298.15, 303.15, 308.15, 313.15, 318.15, 323.15, and 328.15) K. The PEG samples had an average molecular mass (1000 and 10000). The density and refractive index results were correlated to a first-order polynomial with respect to PEG mass fraction at each temperature. It was shown that from the refractive index results the density of the aqueous PEG mixtures can be predicted accurately. The kinematic viscosity results were correlated by a newly modified Eyring's viscosity model. The parameters of the model were determined and presented.

Introduction

A knowledge of the viscosity for polymer−solvent mixtures is required in the polymer process industries. Various models have been presented for predicting the viscosity of polymer−solvent mixtures without recourse to experimental measurements.[1−3] Also, viscosity of polymer−solvent mixtures provides valuable information relating to the size, shape, dimension, structure, degree of polymerization of polymer, and polymer−solvent interactions in the mixture.[4] Unfortunately, viscosity data for their mixtures are scarce.

Poly(ethylene glycols) (PEGs) comprise a series of linear chain polymer of oxyethylene units and due to high water solubility and low toxicity have variety of applications in pharmaceutical, automotive, petroleum, and textile industries.[5−7]

Density and viscosity of PEG aqueous mixtures at limited temperature and polymer concentration ranges have been reported by the other workers.[8−13] In this work, the density, kinematic viscosity, and refractive index of aqueous PEG (molar mass 1000 and 10000) mixtures were measured in the temperature range (298.15 to 328.15) K. The results of density and refractive index measurements were fitted to a first-order polynomial with respect to PEG mass fraction. The experimental kinematic viscosity results were correlated by a newly modified Eyring's viscosity model for polymer−solvent mixtures presented by the authors.[4] The parameters of the model are evaluated and presented.

Experimental Section

Synthesis grade samples of PEG with average molar masses of 1000 (950 to 1050) g/mol and 10000 (9000 to 11250) g/mol were obtained from Merck, Germany. All the

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samples were used without further purification. PEGs were characterized according to their water content determined by Karl Fisher titration using a Metrohm device (KF Coulometer, 684, Switzerland). The water content values obtained were 0.61 and 0.55 mass %, respectively, for PEG 1000 and 10000. They were taken into account when calculating the solution concentrations.

The solutions were prepared by mass, using double distilled water and an analytical balance with ± 0.1 mg accuracy (Shimadzu, model AEU 210). The solutions were slowly stirred in a closed vessel to obtain a true one-phase solution. Then they were put in a thermostated water bath and were allowed to attain thermal equilibrium. The temperature of water bath was measured to the accuracy of ± 0.1K by using a thermometer (Cole Parmer Instrument Company) with subdivisions of 0.1 K.

A 25 cm^3 calibrated glass pycnometer was used for density measurements at (298.15, 303.15, 308.15, 313.15, 318.15, 323.15, and 328.15) K. To calibrate the glass pycnometer, it was filled up with distilled water, and the weight of water was determined (accuracy ± 0.1 mg). Using the reported accurate pure water densities[14] in the temperature range (298.15 to 328.15) K, the volume of pycnometer was determined with uncertainty ± 10^{-4} cm^3. For each solution, four density measurements were performed, and the results were averaged. The uncertainty of density measurements was estimated as ± 10^{-4} g·cm^{-3}.

Kinematic viscosities, ν ($= \eta/\rho$), of the aqueous PEG solutions were measured using three calibrated modified Ostwald viscometer (Cannon-Fenske glass capillary viscometers) with i.d. (0.30, 0.54, and 0.78) ± 2 % mm, respectively for kinematic viscosity ranges (0.5 to 2) × 10^{-6} m^2·s^{-1}, (1.6 to 8) × 10^{-6} m^2·s^{-1}, and (3 to 22) × 10^{-6} m^2·s^{-1}. The flow time was measured with a stop watch capable of recording to 0.01 s. For each solution, the experimental

Table 1. Measured Densities of Aqueous Solutions of (PEG 1000) for Various Mass Fractions (*w*) of PEG and at Different Temperatures

	ρ/g·cm^{-3} at w									
T/K	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40	0.45	0.50
298.15	1.0075	1.0169	1.0250	1.0329	1.0428	1.0510	1.0585	1.0680	1.0785	1.0868
303.15	1.0062	1.0149	1.0237	1.0321	1.0407	1.0505	1.0576	1.0672	1.0778	1.0864
308.15	1.0050	1.0140	1.0221	1.0306	1.0400	1.0484	1.0557	1.0650	1.0742	1.0843
313.15	1.0030	1.0110	1.0192	1.0294	1.0380	1.0454	1.0529	1.0610	1.0702	1.0800
318.15	1.0005	1.0092	1.0175	1.0270	1.0345	1.0421	1.0500	1.0591	1.0662	1.0749
323.15	0.9998	1.0080	1.0149	1.0225	1.0304	1.0395	1.0473	1.0550	1.0645	1.0719
328.15	0.9978	1.0047	1.0128	1.0211	1.0281	1.0351	1.0442	1.0521	1.0606	1.0681

Table 2. Measured Densities of Aqueous Solutions of (PEG 10000) for Various Mass Fractions (*w*) of PEG and at Different Temperatures

	ρ/g·cm^{-3} at w									
T/K	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40	0.45	0.50
298.15	1.0108	1.0186	1.0278	1.0384	1.0466	1.0549	1.0655	1.0737	1.0839	1.0935
303.15	1.0095	1.0159	1.0271	1.0369	1.0447	1.0538	1.0627	1.0732	1.0811	1.0895
308.15	1.0090	1.0150	1.0254	1.0350	1.0438	1.0505	1.0606	1.0698	1.0780	1.0860
313.15	1.0064	1.0143	1.0244	1.0317	1.0407	1.0490	1.0570	1.0677	1.0761	1.0826
318.15	1.0043	1.0120	1.0200	1.0291	1.0373	1.0460	1.0552	1.0639	1.0720	1.0804
323.15	1.0004	1.0102	1.0197	1.0268	1.0344	1.0425	1.0510	1.0612	1.0683	1.0746
328.15	0.9984	1.0069	1.0168	1.0232	1.0313	1.0399	1.0484	1.0570	1.0647	1.0711

Table 3. Coefficients *a* and *b* and the AADs Obtained for Aqueous Solutions of (PEG 1000) from Equation 2[a]

	eq 2			eqs 2, 4, and 5
T/K	a	b	AAD	AAD
298.15	0.9986	0.1751	0.06	0.57
303.15	0.9968	0.1777	0.06	0.44
308.15	0.9962	0.1737	0.04	0.38
313.15	0.9945	0.1690	0.07	0.43
318.15	0.9930	0.1640	0.04	0.46
323.15	0.9912	0.1608	0.09	0.52
328.15	0.9892	0.1571	0.09	0.57

[a] By correlating the experimental densities at various temperatures and the AADs obtained from eq 2 by using the evaluated coefficients *a* and *b* at each temperature (eqs 4 and 5).

Table 4. Coefficients *a* and *b* and the AADs Obtained for Aqueous Solutions of (PEG 10000) from Equation 2[a]

	eq 2			eqs 2, 6, and 7
T/K	a	b	AAD	AAD
298.15	1.0007	0.1843	0.06	0.52
303.15	0.9997	0.1810	0.06	0.41
308.15	0.9993	0.1745	0.07	0.31
313.15	0.9977	0.1720	0.06	0.43
318.15	0.9950	0.1711	0.03	0.31
323.15	0.9935	0.1652	0.09	0.53
328.15	0.9893	0.1677	0.08	0.46

[a] By correlating the experimental densities at various temperatures and the AADs obtained from eq 2 by using the evaluated coefficients *a* and *b* at each temperature (eqs 6 and 7).

viscosity was obtained by averaging 5 to 7 flow time measurements. The uncertainty of kinematic viscosity measurements was estimated as $\pm$ 10^{-9} m^2·s^{-1}.

The viscometers were calibrated by the following equation:

$$\frac{\eta}{\rho} = Kt - \frac{L}{t} \qquad (1)$$

where η is the absolute viscosity, ρ is the density, and t is the flow time of the pure calibration liquid. In this work, by using the absolute viscosity (η) and density (ρ) of pure dimethyl formamide (DMF), the characteristic viscometer constants K and L were obtained.

Refractive indices were measured using a thermostatically controlled Abbe refractometer (Atago 3T, made in Japan). The refractometer was calibrated according to the instruction of the manufacturer by using pure water refractive index at certain temperatures. A minimum of three independent readings were taken for each composition. The uncertainty of refractive index measurements was lower than 0.05 %.

Results and Discussion

Density Model. The obtained density results in Tables 1 and 2 were correlated to a first-order polynomial with respect to PEG mass fraction in the following form:
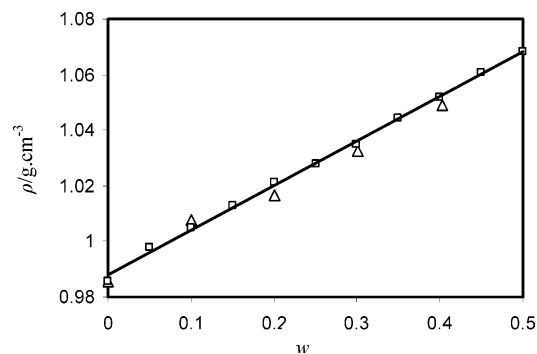
$$\rho/\text{g·cm}^{-3} = a + bw \qquad (2)$$

where ρ is the density, w is the mass fraction of PEG, and

Figure 1. Density ρ of PEG (10000) aqueous solution versus PEG weight fraction w at 328.15 K: $\square$, measured in this work; $\triangle$, measured by Muller and Ramussen.[15] The line indicates the measured densities of this work as correlated by eq 2.

a and b are the coefficients of the above linear equation. Coefficients of eq 2 for aqueous PEG mixtures are presented in Tables 3 and 4, respectively, for PEG 1000 and 10000. The percent average absolute deviation (AAD) was used for comparisons of experimental and correlated results according to the following equation:

$$\text{AAD} = \left[\left(\sum_{i=1}^{n} \left(\frac{y_{\text{exp},i} - y_{\text{cal},i}}{y_{\text{exp},i}} \right) \right) \right] \times \frac{100}{n} \qquad (3)$$

where $y_{\text{exp},i}$, $y_{\text{cal},i}$, and n are experimental point, calculated point, and number of data points, respectively. These

Table 5. Measured Refractive Index (n_D) of Aqueous Solutions of (PEG 1000) for Various Mass Fractions (w) of PEG and at Different Temperatures

T/K	n_D at w									
	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40	0.45	0.50
298.15	1.3394	1.3461	1.3531	1.3600	1.3675	1.3750	1.3821	1.3911	1.3982	1.4060
303.15	1.3388	1.3454	1.3522	1.3593	1.3661	1.3735	1.3810	1.3905	1.3969	1.4051
308.15	1.3379	1.3445	1.3515	1.3586	1.3652	1.3728	1.3805	1.3890	1.3963	1.4035
313.15	1.3373	1.3440	1.3508	1.3575	1.3640	1.3721	1.3796	1.3885	1.3950	1.4029
318.15	1.3366	1.3433	1.3501	1.3565	1.3635	1.3709	1.3787	1.3877	1.3935	1.4012
323.15	1.3350	1.3421	1.3494	1.3560	1.3620	1.3698	1.3770	1.3847	1.3930	1.4002

Table 6. Measured Refractive Index (n_D) of Aqueous Solutions of (PEG 10000) for Various Mass Fractions (w) of PEG and at Different Temperatures

T/K	n_D at w									
	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40	0.45	0.50
298.15	1.3401	1.3470	1.3541	1.3617	1.3700	1.3765	1.3841	1.3915	1.4000	1.4077
303.15	1.3388	1.3460	1.3534	1.3612	1.3679	1.3758	1.3830	1.3900	1.3987	1.4062
308.15	1.3382	1.3452	1.3524	1.3604	1.3663	1.3740	1.3820	1.3890	1.3985	1.4045
313.15	1.3374	1.3443	1.3519	1.3599	1.3657	1.3731	1.3805	1.3882	1.3970	1.4040
318.15	1.3370	1.3435	1.3506	1.3584	1.3650	1.3715	1.3795	1.3872	1.3950	1.4015
323.15	1.3360	1.3425	1.3498	1.3577	1.3640	1.3708	1.3773	1.3852	1.3933	1.4005

Table 7. Coefficients α and β and the AADs Obtained for Aqueous Solutions of (PEG 1000) from Equation 8[a]

T/K	eq 8			eqs 8, 9, and 10
	α	β	AAD	AAD
298.15	1.3309	0.1488	0.04	0.94
303.15	1.3302	0.1480	0.05	0.96
308.15	1.3295	0.1471	0.04	0.97
313.15	1.3288	0.1467	0.05	0.97
318.15	1.3282	0.1445	0.05	1.03
323.15	1.3284	0.1429	0.04	1.02

[a] By correlating the experimental refractive index at various temperatures and the AADs obtained from eq 8 by using the evaluated coefficients α and β at each temperature (eqs 9 and 10).

comparisons indicate that the AAD was less than 0.09. By considering the variation of polynomial constants (a and b) versus temperature, the following simple temperature functionalities are presented for a and b:

for PEG (1000) $\quad a/\text{g·cm}^{-3} = -0.0003T/\text{K} + 1.0939 \quad$ (4)

$$b/\text{g·cm}^{-3} = -0.0007T/\text{K} + 0.3847 \quad (5)$$

for PEG (10000) $\quad a/\text{g·cm}^{-3} = -0.0003T/\text{K} + 1.0991 \quad (6)$

$$b/\text{g·cm}^{-3} = -0.0007T/\text{K} + 0.3975 \quad (7)$$

where T is the temperature. By using the above equations, the maximum AAD as presented in Tables 3 and 4 is 0.57.

Figure 1 shows the measured densities of aqueous PEG (10000) mixtures at 328.15 K and the linear variation of the measured densities versus weight fraction of PEG according to eq 2. Also in this figure, the only experimental data available for the same mixture and at 328.15 K, measured by Muller and Ramussen,[15] are presented. As it is seen from this figure, there is good agreement between the two measurements.

Refractive Index Model. The refractive index result in Tables 5 and 6 were correlated to a first-order polynomial with respect to PEG mass fraction in the following form:

$$n_D = \alpha + \beta \cdot w \quad (8)$$

where n_D is the refractive index, w is the mass fraction,

Table 8. Coefficients α and β and the AADs Obtained for Aqueous Solutions of (PEG 10000) from Equation 8[a]

T/K	eq 8			eqs 8, 11, and 12
	α	β	AAD	AAD
298.15	1.3319	0.1503	0.03	0.75
303.15	1.3310	0.1493	0.02	0.75
308.15	1.3303	0.1477	0.03	0.78
313.15	1.3296	0.1470	0.04	0.78
318.15	1.3291	0.1447	0.03	0.81
323.15	1.3284	0.1429	0.03	0.83

[a] By correlating the experimental refractive index at various temperatures and the AADs obtained from eq 8 by using the evaluated coefficients α and β at each temperature (eqs 11 and 12).

Table 9. Coefficients σ and κ and the AADs Obtained for Aqueous Solutions of (PEG 1000) from Equation 13[a]

T/K	eq 13			eqs 13, 14, and 15
	σ	κ	AAD	AAD
298.15	−0.5667	1.1762	0.05	0.70
303.15	−0.5979	1.1990	0.07	0.62
308.15	−0.5724	1.1798	0.06	0.59
313.15	−0.5331	1.1497	0.11	0.66
318.15	−0.5131	1.1338	0.08	0.71
323.15	−0.4948	1.1192	0.07	0.85

[a] By correlating the experimental density and refractive index at various temperatures and the AADs obtained from eq 13 by using the evaluated coefficients σ and κ at each temperature (eqs 14 and 15).

and α and β are the coefficients of the above linear equation. Coefficients of eq 8 for aqueous PEG mixtures are presented in Tables 7 and 8 for PEG 1000 and 10000, respectively. The comparisons of experimental and correlated refractive index results indicate that AAD was less than 0.05. By considering the variation of polynomial constants (α and β) versus temperature, the following simple temperature functionalities are presented for α and β:

for PEG (1000) $\quad a = 0.0033T/\text{K} - 1.6253 \quad$ (9)

$$\beta = -0.0026T/\text{K} + 1.9993 \quad (10)$$

for PEG (10000) $\quad a = 0.0053T/\text{K} - 2.2051 \quad$ (11)

$$\beta = -0.0041T/\text{K} + 2.4440 \quad (12)$$

Table 10. Coefficients σ and κ and the AADs Obtained for Aqueous Solutions of (PEG 10000) from Equation 13[a]

	eq 13			eqs 13, 16, and 17
T/K	σ	κ	AAD	AAD
298.15	−0.6321	1.2259	0.04	0.41
303.15	−0.6133	1.2118	0.06	0.48
308.15	−0.5619	1.1737	0.08	0.39
313.15	−0.5489	1.1632	0.06	0.41
318.15	−0.5759	1.1819	0.03	0.47
323.15	−0.5597	1.1689	0.08	0.46

[a] By correlating the experimental density and refractive index at various temperatures and the AADs obtained from eq 13 by using the evaluated coefficients σ and κ at each temperature (eqs 16 and 17).

Table 11. Measured Kinematic Viscosities (ν) of Aqueous Solution of (PEG 10000) for Various Mass Fractions (w) of PEG and at Different Temperatures

	$\nu/10^{-6}$ m^2·s^{-1} at w			
T/K	0.05	0.10	0.15	0.20
298.15	3.3416	5.7560	11.3730	21.9900
303.15	2.9378	5.2710	9.9800	20.1000
308.15	2.5723	4.7122	8.5840	18.0000
313.15	2.2661	4.2593	7.4600	16.4800
318.15	2.0161	3.9000	6.7400	15.0300
323.15	1.8015	3.5210	5.8700	13.6900
328.15	1.6291	3.2730	5.1730	12.3950

By using the above equations for α and β, the maximum AAD as shown in Tables 7 and 8 is 1.03.

By considering the density and refractive index results for the mixtures, the following linear equation is proposed:

$$\rho/\text{g·cm}^{-3} = \sigma + \kappa n_D \tag{13}$$

where ρ is the density and n_D is the refractive index and σ and κ are the coefficients of the above linear equation. Coefficients of eq 13 for aqueous PEG mixtures are presented in Tables 9 and 10 for PEG 1000 and 10000, respectively. The comparisons of experimental and correlated refractive index results indicate that AAD was less than 0.11. By considering the variation of polynomial constants (σ and κ) versus temperature, the following simple temperature functionalities are presented for σ and κ:

for PEG (1000)

$$\sigma = 0.0037T/\text{K} - 1.7029 \tag{14}$$

$$\kappa = -0.0029T/\text{K} + 2.0639 \tag{15}$$

for PEG (10000)

$$\sigma = 0.0028T/\text{K} - 1.4427 \tag{16}$$

$$\kappa = -0.0022T/\text{K} + 1.8682 \tag{17}$$

By using the above equations, the maximum AAD as presented in Tables 9 and 10 is 0.85. Therefore, without recourse to the tedious and time-consuming density measurements, the densities of the studied mixtures, required

in the viscosity model, can be calculated accurately by using eq 13 from the measured refractive index results.

Viscosity Model. The measured kinematic viscosities of aqueous PEG mixtures at different temperatures and concentrations are presented in Tables 11 and 12. The viscosity data are correlated by a newly modified Eyring's viscosity model for water (1) + PEG (2) binary mixtures in the following form:[4]

$$\eta = (2\pi RT)^{1/2}[M_1^{x_1/2}M_2^{x_2/2}/V_1^{x_1}V_2^{x_2}]e^{E_{am}}/RT \tag{18}$$

where x, M, R, T, V, and E_{am} represent respectively mole fraction, molar mass, gas constant, temperature, molar volume, and molar activation energy of the mixtures.

The molar activation energy of mixture (E_{am}) is expressed as:

$$E_{am} = a_m + b_m T \tag{19}$$

where a_m and b_m are mixture parameters. At low concentration of PEG in the binary mixture of water (1) + PEG (2) the following approximation is valid:

$$V_1^{x_1}V_2^{x_2} \approx V_1 = V \tag{20}$$

since, by considering the higher molecular weight of PEGs as compared with that of water, the mole fraction of PEGs in the studied mixtures is very small ($x_2 \approx 0$) and then according to eq 20 the molar volume of mixtures (V) can be used in the kinematic viscosity calculations. Therefore, the final form of viscosity model (eq 18) for a the studied binary mixture of water (1) + PEG (2) will be presented as:

$$\nu = \eta/\rho = (2\pi RT)^{1/2}[M_1^{(x_1/2)-1}M_2^{x_2/2}] \exp[(a_m + b_m T)/RT] \tag{21}$$

where ν is the kinematic viscosity. Equation 21 is rearranged in the following linear form:

$$\ln(\nu/T^{1/2}) = A(a_m + b_m T)/RT \tag{22}$$

where

$$A = \ln[(2\pi R)^{1/2}M_1^{(x_1/2)-1}M^{x_2}/2] \tag{23}$$

Then eq 21 can be presented in the following simple form:

$$\ln(\nu/T^{1/2}) = \alpha_m/T + \beta_m \quad \text{and } \alpha_m = Aa_m/R,$$
$$\beta_m = Ab_m/R \tag{24}$$

According to eq 24, the variation of $\ln(\nu/T^{1/2})$ versus $1/T$ is linear. Equation 24 was used to correlate the measured kinematic viscosities of water (1) + PEG (2) mixtures and to evaluate the parameters α_m and β_m. The results are given in Tables 13 and 14 for PEG 1000 and 10000,

Table 12. Measured Kinematic Viscosities (ν) of Aqueous Solution of (PEG 1000) for Various Mass Fractions (w) of PEG and at Different Temperatures

	$\nu/10^{-6}$ m^2·s^{-1} at w									
T/K	0.05	0.10	0.15	0.20	0.25	0.30	0.35	0.40	0.45	0.50
298.15	1.2581	1.6358	2.2121	3.3014	4.7214	6.3015	7.9258	9.8750	11.8565	13.8525
303.15	1.1122	1.5012	2.0158	3.0015	4.4123	5.7895	7.2691	9.1582	11.1598	13.0158
308.15	0.9825	1.3125	1.7253	2.6890	3.8522	5.1254	6.7894	8.5556	10.4890	12.2581
313.15	0.8356	1.1625	1.5282	2.3693	3.4585	4.7052	6.2895	7.9258	9.8563	11.5861
318.15	0.7235	1.0021	1.3584	2.0514	3.0158	4.1981	5.7896	7.3581	9.2581	10.8964
323.15	0.6015	0.8872	1.1258	1.8122	2.6581	3.6983	5.2582	6.7582	8.6582	10.2583
328.15	0.5106	0.7689	1.0025	1.5986	2.3596	3.3895	4.8251	6.1584	8.0112	9.6582

Table 13. Parameters of Kinematic Viscosity Model, Equation 24, for Aqueous (PEG 1000) Mixtures and the Standard Deviations (SD)

w	α_m	β_m	SD	w	α_m	β_m	SD
0.05	3113.8	−13.022	0.033	0.30	2220.6	−8.437	0.031
0.10	2659.0	−11.238	0.030	0.35	1758.7	−6.667	0.030
0.15	2793.1	−11.388	0.034	0.40	1674.2	−6.161	0.031
0.20	2567.6	−10.234	0.034	0.45	1418.7	−5.124	0.033
0.25	2486.2	−9.597	0.029	0.50	1327.2	−4.668	0.025

Table 14. Parameters of Viscosity Model, Equation 24, for Aqueous (PEG 10000) Mixtures and the Standard Deviations (SD)

w	α_m	β_m	SD
0.05	2518.6	−10.092	0.012
0.10	2037.2	−7.9253	0.014
0.15	2719.9	−9.5372	0.015
0.20	2020.9	−6.5292	0.013

respectively. The standard deviations (SD) were calculated to determine quantitatively the accuracy of the proposed model by using the following equation:

$$SD = \left[\frac{\sum_{i=1}^{n}(y_{exp,i} - y_{cal,i})^2}{n - p}\right]^{1/2} \quad (25)$$

where $y_{exp,i}$, $y_{cal,i}$, n, and p are experimental point, calculated point, number of data points, and number of adjusted parameters, respectively. SDs reported in Tables 13 and 14 indicate the high quality of fitting according to eq 24.

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