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Studies on the Diffusion Coefficients of Amino Acids in Aqueous Solutions

Youguang Ma,* Chunying Zhu, Peisheng Ma, and K. T. Yu

School of Chemical Engineering and Technology, State Key Laboratory of Chemical Engineering,
Tianjin University, Tianjin 300072, PR China

The diffusion of amino acids in aqueous solution was investigated experimentally by a holographic interferometric technique where the real-time holographic interference fringes indicating the concentration profiles of the liquid were obtained by an automatic photographing and memorizing program. The reliability of the instrument was verified by the measurement of the diffusion coefficient of KCl and sucrose in aqueous solution at 298.15 K. Furthermore, the diffusion coefficients of glycine, L-alanine, L-valine, L-isoleucine, L-serine, L-threonine, and L-arginine in aqueous solution at 298.15 K were measured, and the affecting factors of molecular structure and polarity were analyzed and discussed. In addition, on the basis of the Gordon model of the diffusion coefficient in the literature, a new semiempirical model was proposed to predict the liquid diffusion coefficients of amino acids in aqueous solutions. The equation parameters were fitted by the experimental data of seven amino acids in this work and four in the literature.

Introduction

The diffusion coefficient is very important basic data for the application of chemical and equipment design involving mass-transfer processes. However, there is limited experimental data of liquid diffusion coefficients in the literature.[1−4] Therefore, the experimental investigation, correlating experimental data, and development of a prediction model are of practical and theoretical significance. Although amino acids are among the simplest biochemicals, they have many similarities to more complex biomolecules such as antibiotics that are used widely as nutrients and medicines,[5−7] so the study of their diffusivity in aqueous solution is of fundamental importance.[8−10]

In this article, the diffusion of amino acids in aqueous solution is studied by means of a real-time holographic interferometer. A new semiempirical model is proposed to predict the liquid diffusion coefficients of amino acids in aqueous solutions.

Experimental Section

Holographic interferometry has been widely applied to quantitative measurements of various physical parameters in fluids such as velocity, temperature, concentration, density, and so forth with the advantages of high sensitivity and accuracy, a large capacity for information, nonintrusiveness, the capability of making instantaneous measurements, and the visualization of the overall process.[11]

The experimental setup is depicted in Figure 1. A transparent rectangular cell that holds the diffusing liquids (dimensions 10 mm × 15 mm × 100 mm) was filled through a capillary tube from the bottom to minimize turbulence and mixing. The less dense fluid was put into the cell first, and then the more dense liquid was admitted slowly from the bottom to displace the first liquid. The liquids were allowed to diffuse for about 30 min before taking holograms. This time delay is required to minimize turbulence at the interface of the two liquids.

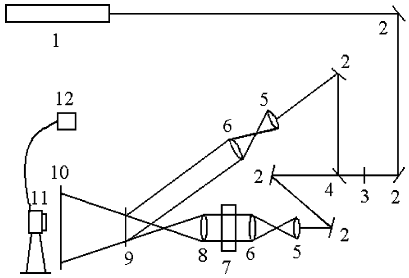

Figure 1. Optical representation of the laser holographic interferometer. 1, He−Ne laser; 2, mirors; 3, shutter; 4, beam splitter; 5, spatial filters; 6, lenses; 7, simulator; 8, amplifying lens; 9, holographic plate; 10, frosted glass; 11, CCD camera; 12, computer.

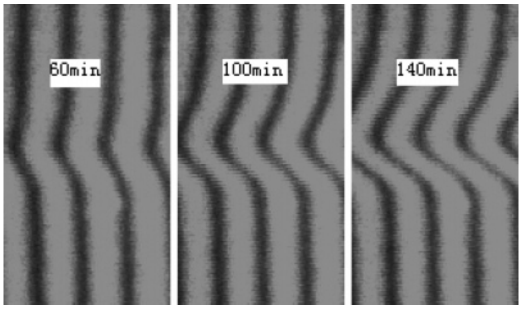

Figure 2. Holographic interference stripes at different times.

The interferential fringes were photographed by a CCD with 795 × 795 pels and retained in the computer by an image board. An automatic photographing and memorizing program was used to improve the precision of the time register. Holographic interference fringes at different times are shown in Figure 2.

Holographic interferometry records the change in the index of refraction. Because the refractive index of a small concentration interval solution varies linearly with the concentration C of the solute, this method can be used to determine changes in the concentration profile.

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* Corresponding author. E-mail: ygma@tju.edu.cn. Tel: +86-22-27404772. Fax: +86-22-27404757.

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Table 1. Comparison of Diffusion Coefficients between the Experiment and the Literature

	C/mol ·L^{-1}	$10^{10}D_{\text{expt}}$/m^2 ·s^{-1}	$10^{10}D_{\text{lit}}$/m^2·s^{-1}	ref	AD^a/%
KCl	0.33	1.83	1.831	13	0.05
sucrose	0.1	4.89	4.87	14	0.41
	0.3	4.27	4.26	14	0.23
	0.5	3.67	3.68	14	0.27
	0.7	3.06	3.04	14	0.66

[a] AD is the abbreviation for average deviation.

To facilitate this method, the diffusion process is treated as one-dimensional and starts from the interface between two liquids with concentrations $C_1$and C_2. On the basis of Fick's law and the concentration distribution given by Crank,[12] the diffusion coefficient is given by

$$D = \frac{x^2}{4\Delta t}\{[\text{erfinv}(Ct_2)]^{-2} - [\text{erfinv}(Ct_1)]^{-2}\} \tag{1}$$

where

$$Ct = \frac{[2C(x, t) - (C_1 + C_2)]}{(C_2 - C_1)}$$

Δt is the time interval between two exposures, erfinv is the inverse function of erf, and $C(x, t)$ is the solution concentration at section x and time t.

Amino acid (Tianjin Amino Acid Co., China; mass fraction >0.997), KCl, and sucrose (Tianjin No. 1 Chemical Plant, China; mass fraction >0.999) were used in this work. The samples for the diffusion coefficients were prepared by weight (FA2004N balance with an accuracy of $\pm 1 \times 10^{-4}$ g) for the whole mole concentration range; the concentration difference used between the less dense and the more dense fluid is 0.04 mol ·L^{-1}, the system temperature was kept at (298.15 ± 0.01) K, and every measurement was repeated three times. The system was validated by the KCl and sucrose aqueous solution diffusion at 298.15 K,[13,14] which is reliable and commonly accepted. The diffusion coefficients of KCl and the sucrose aqueous solution are given in Table 1; the average deviation is less than 0.7%. The experimental uncertainty in the diffusion coefficient is approximately $\pm 1 \times 10^{-12}$ m^2·s^{-1}.

Results and Discussions

The diffusion coefficients of glycine, L-alanine, L-valine, L-isoleucine, L-serine, L-threonine, and L-arginine in aqueous solution at 298.15 K were measured, and the experimental results are shown in Table 2.

At the same concentration, the diffusion coefficients of amino acids vary in the order D(glycine) > D(alanine) > D(serine) > D(threonine) > D(valine) > D(isoleucine) > D(arginine), which can be explained from two aspects. At first, the diffusion coefficient is influenced by solute size. On the basis of the simplest Stokes−Einstein equation, $D_{\text{AB}} = kT/(6\pi\eta_0 r)$, the smaller the molecular volume of the diffusing species, the larger the diffusion coefficient. Upon inspecting the molecular constitution of amino acid, it can be found that the side chains of alanine, valine, and isoleucine are nonpolar and their molecular chains are longer and longer in turn, which is also true for serine, threonine, and arginine, and their molecular volume gradually increases; therefore, their diffusivities are smaller and smaller.

Second, the diffusion coefficient can be influenced by the molecular polarity because the solute polarity can result in electrostriction. In this experiment, the solvent is water, so the polar solute molecule combines adjacent water molecules to produce larger aggregates, which leads to an increase of the effective diffusive volume and a decrease of the diffusion coefficient. In aqueous solution, alanine, valine, and isoleucine with hydrophobic side chains can combine only a small quantity of water molecules. Serine and threonine are polar amino acids with hydrophilic side chains and no charge, whereas arginine is a polar amino acid with a positive charge, which leads to an increase in the hydrated volume. Taking into account the hydrophobic and hydrophilic effects of the side chains simultaneously, the strength of interaction between the solute and water remains in the order glycine (or other unpolar amino acids) < threonine (or serine) < arginine. Hence, the diffusivities gradually decrease with increasing interactive strength.

In view of the complex structure of the liquid and the diffusion process, no precisely theoretical equation for the prediction of binary liquid diffusion coefficients has been developed. The Stokes−Einstein equation is a classically theoretical model of the diffusion coefficient based on hydrodynamic theory

$$D_{\text{AB}} = \frac{kT}{6\pi\eta_{\text{B}}r_{\text{A}}} \tag{2}$$

where D_{AB} is the diffusion coefficient, r_{A} is the molecular radius of solute A, and η_{B} is the viscosity of solvent B. Equation 2 is applied to the diffusion process of spherical molecules in dilute solution.

On the basis of the Eyring absolute velocity theory, an approach for estimating diffusivity was developed[15]

$$D_{\text{AB}} = \frac{\delta^2}{V_{\text{f}}^{1/3}}\left(\frac{RT}{2\pi M}\right)^{1/2} \exp\left(-\frac{E_{\text{AB}}}{RT}\right) \tag{3}$$

where δ is the characteristic distance between two adjacent molecules, V_{f} is the free molecular volume, M is the molecular weight of the solution, and E_{AB} is the diffusion energy of activation. Equation 3 is mostly used with the diffusion process of a gas in a liquid.

According to statistical mechanics theory, Bearman[16] presented a prediction model

$$D_{\text{AB}} = \frac{kTv}{\xi_{\text{AB}}}\frac{\text{d} \ln a_{\text{B}}}{\text{d} \ln C_{\text{B}}} \tag{4}$$

where v is the molecular motion velocity, ξ_{AB} is the frictional coefficient between components A and B, a_{B} is the activity of solute B, and C_{B} is the molar concentration of solute B.

In addition, Hayduk and Laudie[17] correlated the molar volumes of organic compounds with the aqueous diffusion coefficient, and an empirical equation was proposed

$$D_{\text{w}} = \frac{13.26 \times 10^{-5}}{{\eta_{\text{w}}}^{1.4} \times {V_{\text{A}}}^{0.589}} \tag{5}$$

where D_{w} is the diffusion coefficient of the organic com-

Table 2. Comparison of the Diffusion Coefficient between the Experimental and Calculated Values

C	$10^2\eta$	$10^{10}D_{\text{exptl}}$	$10^{10}D_{\text{eq 10}}$	AD	$10^{10}D_{\text{Gor}}$	AD	C	$10^2\eta$	$10^{10}D_{\text{exptl}}$	$10^{10}D_{\text{eq 10}}$	AD	$10^{10}D_{\text{Gor}}$	AD
mol·L^{-1}	Pa·s	$\text{m}^2\cdot\text{s}^{-1}$	$\text{m}^2\cdot\text{s}^{-1}$	%	$\text{m}^2\cdot\text{s}^{-1}$	%	mol·L^{-1}	Pa·s	$\text{m}^2\cdot\text{s}^{-1}$	$\text{m}^2\cdot\text{s}^{-1}$	%	$\text{m}^2\cdot\text{s}^{-1}$	%
Glycine													
0.1057	1.0169	10.40	10.41	0.10	10.44	0.38	0.7064	1.1182	9.57	9.57	0.00	9.51	0.63
0.3056	1.0506	10.01	9.98	0.30	10.11	1.00	0.9045	1.1517	9.43	9.42	0.11	9.24	2.01
0.5045	1.0843	9.73	9.75	0.21	9.80	0.72							
L-Alanine													
0.0997	1.0317	9.04	9.04	0.00	8.96	0.88	0.7002	1.2222	8.12	8.12	0.00	7.57	6.77
0.2996	1.0955	8.59	8.59	0.00	8.44	1.75	0.9006	1.2857	7.92	7.92	0.00	7.20	9.09
0.4996	1.1587	8.34	8.34	0.00	7.98	4.32							
L-Valine													
0.1000	1.0607	7.12	7.13	0.14	7.37	3.51	0.4000	1.2429	6.45	6.44	0.16	6.27	2.79
0.2000	1.1215	6.79	6.77	0.29	6.96	2.50	0.5001	1.3036	6.33	6.33	0.00	5.97	5.69
0.2999	1.1822	6.55	6.57	0.31	6.60	0.76							
L-Serine													
0.0100	1.0032	9.16	9.16	0.00	9.18	0.22	0.0700	1.0207	8.71	8.71	0.00	9.03	3.67
0.0301	1.0089	8.91	8.91	0.00	9.13	2.47	0.0902	1.0267	8.65	8.65	0.00	8.98	3.82
0.0500	1.0148	8.79	8.79	0.00	9.08	3.30							
L-Threonine													
0.1006	1.0431	7.86	7.86	0.00			0.4002	1.1724	7.25	7.26	0.14		
0.2002	1.0862	7.58	7.58	0.00			0.5003	1.2155	7.15	7.14	0.14		
0.2994	1.1293	7.42	7.41	0.13									
L-Arginine													
0.1002	1.0889	7.01	7.01	0.00			0.4001	1.3554	6.21	6.19	0.32		
0.2000	1.1777	6.61	6.61	0.00			0.5000	1.4443	6.04	6.05	0.17		
0.2949	1.2666	6.36	6.37	0.16									
L-Isoleucine													
0.0101	1.0075	7.61	7.61	0.00			0.0399	1.0301	7.32	7.32	0.00		
0.0203	1.0153	7.50	7.49	0.13			0.0500	1.0376	7.25	7.25	0.00		
0.0301	1.0225	7.39	7.40	0.14									
β-Alanine[23]													
0.0281	1.0064	9.36	9.36	0.00			1.7660	1.5630	7.79	7.88	1.16		
0.0288	1.0065	9.35	9.36	0.11			2.3397	1.8467	7.52	7.53	0.13		
0.0299	1.0068	9.34	9.35	0.11			2.6648	2.0400	7.40	7.33	0.95		
0.0344	1.0078	9.33	9.33	0.00			3.0083	2.2748	7.20	7.13	0.97		
0.1110	1.0326	9.18	9.12	0.65			3.9313	3.1133	6.63	6.58	0.75		
0.5605	1.1401	8.67	8.67	0.00			4.5431	3.9032	6.25	6.22	0.48		
0.5886	1.1478	8.68	8.65	0.35			5.4194	5.5648	5.60	5.70	1.79		
1.1225	1.3130	8.18	8.28	1.22									
α-Amino-*n*-butyricacid[24]													
0.20	1.073	8.00	8.01	0.12			0.80	1.329	7.20	7.21	0.14		
0.40	1.150	7.72	7.72	0.00			1.00	1.437	6.97	6.97	0.00		
0.60	1.234	7.45	7.46	0.13									
α-Alanine[24]													
0.0271	1.0076	9.11	9.11	0.00			0.6786	1.1995	8.27	8.27	0.00		
0.0281	1.0076	9.11	9.11	0.00			0.9025	1.2773	8.02	8.03	0.12		
0.2233	1.0596	8.84	8.82	0.23			1.1948	1.3876	7.72	7.73	0.13		
0.4374	1.1223	8.56	8.55	0.12			1.5406	1.5309	7.41	7.39	0.27		
Urea[25]													
0.1250	1.0053	13.73	13.79	0.44			1.0000	1.0804	13.07	13.07	0.00		
0.2475	1.0118	13.63	13.60	0.22			1.5000	1.1532	12.74	12.79	0.39		
0.5000	1.0294	13.43	13.37	0.45			2.0000	1.2476	12.45	12.51	0.48		
0.7500	1.0522	13.26	13.21	0.38			3.0002	1.5016	11.89	11.94	0.42		
0.9782	1.0777	13.09	13.08	0.08			3.9999	1.8424	11.43	11.37	0.52		

pound in aqueous solution, η_w is the viscosity of water at 298.15 K, and V_A is the molar volume of the organic compound.

Wilke and Chang[18] developed an aqueous diffusion coefficient correlation for diffusion in associated liquids such as water

$$D_w = \frac{7.4 \times 10^{-8} \times (2.6M_w)^{0.5} \times T}{\eta_w \times V_A^{0.6}} \tag{6}$$

where M_w is the molecular weight of the water. Equation 6 is widely employed to estimate the diffusion coefficient of a small-molecule solute in dilute aqueous solution.

Gordon[19] proposed a predictive model of the diffusion coefficient that involves the change in the thermodynamic potential of the solute with concentration

$$D = D_0\frac{\eta_0}{\eta}\left(1 + C_A\frac{\partial \ln \gamma_A}{\partial C_A}\right) \tag{7}$$

where D_0 is the diffusion coefficient at $C = 0$, γ_A is the activity coefficient of solute A, C_A is the molar concentration of solute A, and η_0 and η are the viscosities of the solvent and solution, respectively. The Gordon model is widely used to calculate the diffusion coefficients of the aqueous solutions of nonelectrolytes

Because of the lack or difficulty of attaining some necessary model parameters for amino acids in eqs 2 to 6, it is very difficult to calculate diffusivity directly through these equations. The Gordon model needs only the viscosity

Table 3. Parameters Used to Calculate Aqueous Diffusion Coefficients for Different Aqueous Solutions at 298.15 K

system	$10^{10}\alpha/\text{m}^2\cdot\text{s}^{-1}$	β	ω	κ
glycine	6.35950	−0.306423	0.542003	−0.0769850
L-alanine	8.91305	−0.478053	0.00142255	−1.32810
L-vanine	3.37618	−0.0409380	0.814857	−0.136554
L-isoleucine	6.71071	−0.991084	0.0867076	−0.108767
L-serine	8.34458	−0.630560	0.0271490	−0.284055
L-threonine	2.19184	−0.235107	2.18267	−0.0525062
L-arginine	3.55843	−0.136856	0.711910	−0.144486
β-alanine[23]	11.3080	−0.237018	−0.215559	0.0678537
α-amino-*n*-butyricacid[24]	8.41313	−0.257490	−0.0900810	0.673494
α-alanine[24]	9.15850	−0.246708	−0.0760892	0.745259
urea[25]	3.30371	−0.213478	3.04353	−0.0244751

and relation of the activity coefficient to the molar concentration of the solute, which can be easily obtained by experimental measurement or theoretical estimation. In this work, the Gordon model is further developed and modified.

In the Margules equation, the activity coefficient of the solute is expressed as

$$\ln \gamma_A = [A_{AB} + 2(A_{BA} - A_{AB})x_A]x_B^{\,2} \tag{8}$$

where A_{AB} and A_{BA} are model parameters and x_A and x_B are the molar fractions of solute and solvent, respectively.

For the amino acid of very low solubility, $x_A \ll 1$ and $x_B \approx 1$, the total molar concentration of the solution C_t is approximately constant, with

$$\frac{\partial \ln \gamma_A}{\partial C_A} = \frac{1}{C_t}\frac{\partial \ln \gamma_A}{\partial x_A}$$

Equation 7 can be rewritten as

$$D = D_0\frac{\eta_0}{\eta}(1 + \omega C_A) \tag{9}$$

$\omega = 2(A_{BA} - A_{AB})/C_t$ is a new model parameter.

Research[17,20−22] shows that it could be more reasonable that the diffusion coefficient is exponentially related to viscosity and concentration; consequently, a new semiempirical model was proposed to predict the liquid diffusion coefficients at constant temperature

$$D = \alpha\left(\frac{\eta_0}{\eta}\right)^{\beta}(1 + \omega C_A^{\,\kappa}) \tag{10}$$

where α, β, κ, and ω are adjustable parameters.

The equation parameters were fit by the experimented data of seven amino acids in this work and four in the literature as shown in Table 3, and the comparison of prediction results of diffusivities by eq 10 with the Gordon model is shown in Table 2, where D_0 is obtained by extrapolating the curve of diffusivity to concentration; the activity coefficients at different concentrations are obtained from the literature.[26] It can be observed from Table 2 that the total average percent deviation between the model prediction values by eq 10 and the experimental values is less than 0.6%; the present model is better than the Gordon model.

Conclusions

A real-time holographic interferometer and a simulator for liquid−liquid diffusion were specially designed and constructed in order to measure the liquid diffusivity of transparent fluids. The diffusion coefficients of glycine, L-alanine, L-valine, L-isoleucine, L-serine, L-threonine, and L-arginine in aqueous solution at 298.15 K were measured. The diffusion coefficients of amino acids decrease with increasing volume and polarity of the diffusing aggregate in dilute aqueous solutions.

A semiempirical model for estimating the diffusion coefficients of amino acids in aqueous solution is proposed, and the average deviation between model prediction values and experimental values is less than 0.6%. The present model is better than the Gordon model.

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