Determination of Binary Gas Diffusion Coefficients Using Digital Holographic Interferometry

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On the basis of the Mach−Zehnder optical interference model, a digital holographic interferometric experimental system was set up, and a new diffusion cell especially for measuring the binary gas diffusion coefficient was designed and constructed. With the theory for measuring the binary gas diffusion coefficient and the interference fringe processing method introduced before, the binary gas diffusion coefficient can be obtained. In comparison with the reference data of O_2 in air, the experiment accuracy of the system was verified. The diffusion coefficients of H_2, He, NH_3, CH_4, O_2, and CO_2 in air at $T = (278.15 \text{ to } 343.15)$ K under normal atmospheric pressure were measured and compared with results of a correlation. The comparison showed that the results are reasonable.

Introduction

Binary gas diffusion coefficients as an important thermophysical parameter are widely used in industrial production, chemical engineering equipment design, control of atmospheric pollution, and many other fields. Also, it is concerned with the separation and absorption of harmful gases in the absorption tower, the mixing of gasified fuel with air in the chamber of internal combustion engine, and the dispersion of automobile exhaust in atmosphere, and so forth. Hence, accurate and reliable data of binary gas diffusion coefficients are very important. However, compared with the other thermophysical properties of the gas phase, the diffusion coefficients are less than those reported in the literature. The main methods of obtaining the binary gas diffusion coefficient include calculation using a correlation, numerical simulation, and experimental measurement. Currently, the empirical correlations used to calculate the binary gas diffusion coefficient are limited to a narrow range of applications and low accuracy. The numerical simulation is based on simple fluid models, and the accuracy is also not good. So, the precise experimental method is still an indispensable way for obtaining the accurate binary gas diffusion coefficient.

Currently, the experimental methods for measuring binary gaseous diffusion coefficients mainly include the following: diaphragm cells,[1] Taylor's dispersion,[2] dynamic light scattering,[3,4] gas chromatography,[5] and capillary[6] and holographic interferometry.[7] Compared with other methods, digital real-time holographic interferometry has many advantages such as nondirect contact with measured fluids and high accuracy of measurement, and it can monitor the change of parameters during the entire experimental process directly. It is a widely used experimental method for measuring thermophysical properties of fluids and has been used by many researchers to measure the diffusion coefficient of liquids; its accuracy and reliability have been verified.[8,9]

On the basis of our previous work,[11,12,14] a holographic interferometric experimental system for measuring the binary gas diffusion coefficient was used. A diffusion cell was designed and constructed to achieve the process of gas diffusion. Then the uncertainty of the experimental system was analyzed, and the reliability of the system was verified. The binary diffusion coefficients of H_2, He, NH_3, CH_4, O_2, and CO_2 in air at $T = (278.15 \text{ to } 343.15)$ K under normal atmospheric pressure were measured.

Figure 1. Interference fringe image of t_0, t_1, and t_2.

Measurement Theory

Binary gaseous diffusion in a narrow vertical diffusion cell can be seen as one-dimensional diffusion. In the experiment, along with the progress of diffusion, the concentration of the gas changes continuously. These changes include the change of refractive index and the change of object wave's phase. They all lead to the continuous change of interference fringes. The interference fringes as a function of time are shown in Figure 1. By processing these interference fringes, the difference of the object wave's phase can be obtained. With the relationship between the difference of object wave's phase and the diffusion coefficient we can calculate the diffusion coefficient.

The basic theory of the experiment is Fick's second law of one-dimension diffusion, which can be written as[10]

$$\frac{\partial c}{\partial t} = D_{AB}\frac{\partial^2 c}{\partial z^2} \quad (1)$$

The final equation for calculating D_{AB} can be deduced as[12]

$$D_{AB} = \Delta z_m{}^2 \frac{t_1/t_2 - 1}{8t_1 \ln(t_1/t_2)} \quad (2)$$

where D_{AB} is the binary diffusion coefficient, t_1 and t_2 are the moments of collecting the interference fringes which may be

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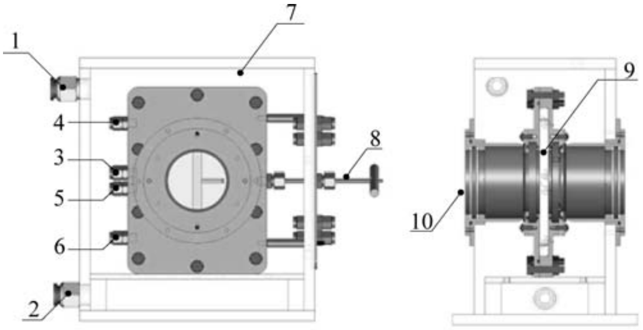

Figure 2. Diffusion cell subsystem of binary gas.

read from the computer timer, and Δz_m is the vertical distance between the two extreme points of the solution's concentration difference which can be extracted from the interference fringe image by image processing methods. The detailed experimental theory and interference fringe processing method have been described previously.[12]

Experimental Section

Experimental Apparatus. The experimental system used in this work is a digital holographic interferometric system. The system includes three parts: a Mach−Zehnder optical interference subsystem, the thermostatic water bath subsystem, and the diffusion cell subsystem. The first two parts were reported in our previous work.[11]

For measuring binary gas diffusion, a new diffusion cell subsystem was designed and constructed as shown in Figure 2. It includes two parts: a diffusion cell and the perspex water tank. To make sure that the experiment goes on in a stable temperature environment, the diffusion cell is fixed in the perspex water tank by the two flanges on both sides of the tank, and the perspex water tank is supplied by an additional thermostatic water system whose temperature can be controlled from (273.15 to 353.15) K with the uncertainty of ± 0.2 K.[14]

The injection of gas is done as follows: First, the baffle plate was closed, and the inlet and outlet for the upside gas were opened. Then, the first gas is injected slowly through the inlet, and with the injection of the first gas, air was ejected out. After a while, when the gas flow is stable, the outlet and inlet were closed, and the injection of upside gas was finished. The injection of the underside gas was the same. When the injection of the two sides of diffusion cell was complete, the draw rod was removed, and the collection of interference fringe began.

Table 1. Experimental Uncertainties of Temperature and the Mass Diffusion Coefficient

temperature	platinum resistance thermometer, u_1	±0.01 K
	data collection and process detector equipment, u_2	±0.025 K
	temperature control system, u_3	±0.05 K
	temperature stability of constant temperature cabinet, u_4	±0.05 K
	combined standard uncertainty, u_c	±0.08 K
mass diffusion coefficient	time of getting hologram, u_1	$\pm 2.22 \cdot 10^{-6}$
	distance between two extreme points of concentration, u_2	±0.1 %
	combined standard uncertainty, u_c	±0.1 %

Experimental Verification and Uncertainty Analysis. The mass diffusion coefficient of O_2 in air under standard conditions was measured to verify the accuracy and reliability of the system. Figure 3 showed the interference fringe image results. By processing the interference fringe images as a function of time, the mean value of the mass diffusion coefficient of O_2 in air is obtained. Comparing the experimental results with literature values,[13] the average of the relative deviations is within 0.4 %, and the experimental system is verified to be reliable.

The experimental uncertainties of temperature and the diffusion coefficient are estimated to be no greater than ± 0.16 K and ± 0.2 %, respectively,[14] as shown in Table 1.

Results and Discussion

The binary diffusion coefficients of H_2, He, NH_3, CH_4, O_2, and CO_2 in air at $T = (278.15 \text{ to } 343.15)$ K under normal atmospheric pressure were measured. For the samples of the different gases, the mass fraction purity of each gas was better than 0.999. The measured results and their uncertainties at each experimental state point are listed in Tables 2 and 3.

For comparison, the Fuller correlation which has been proved to be a quite reliable correlation[13] was used to calculate the diffusion coefficient of the studied gas in air within the same temperature and pressure conditions. The Fuller correlation is

$$D_{AB} = \frac{0.00143 T^{1.75}}{p M_{AB}^{1/2} [(\sum v)_A^{1/3} + (\sum v)_B^{1/3}]^2} \tag{3}$$

where T is absolute temperature, p is pressure, and M_{AB} is mean molar mass, where $M_{AB} = 2[(1/M_A) + (1/M_B)]^{-1}$. M_A and M_B are the molar masses of gas A and gas B, and $\sum v$ is atomic diffusion volume and can be obtained from specific tables.[13]

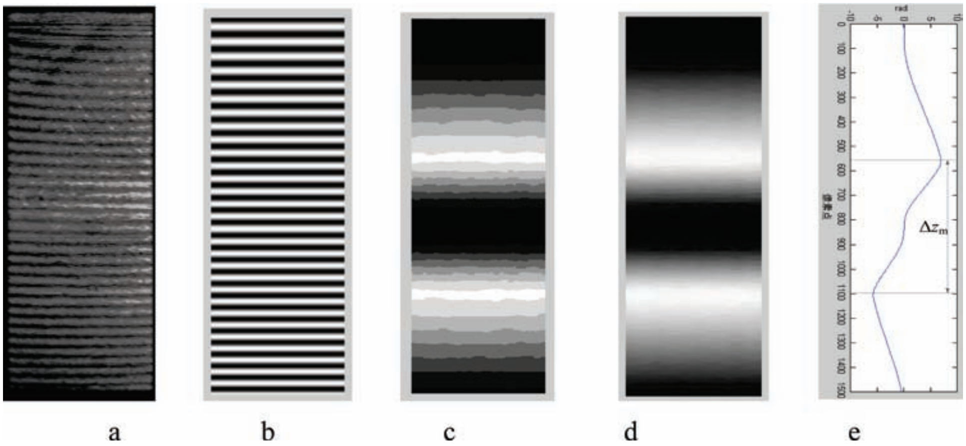

Figure 3. Interference fringe image processing results.

Table 2. Measured Mass Diffusion Coefficients D_{AB} of H_2, He, and NH_3 in Air from T = (278.15 to 343.15) K under Normal Atmospheric Pressure

	H_2 (1) + air (2)		He (1) + air (2)		NH_3 (1) + air (2)	
T	D_{AB}	$u_{D_{12}}$	D_{AB}	$u_{D_{12}}$	D_{AB}	$u_{D_{12}}$
K	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$
278.15	0.697	±0.0025	0.645	±0.0024	0.231	±0.0025
283.15	0.721	±0.0025	0.662	±0.0024	0.238	±0.0024
288.15	0.744	±0.0024	0.680	±0.0024	0.245	±0.0025
293.15	0.767	±0.0025	0.697	±0.0025	0.254	±0.0024
298.15	0.791	±0.0025	0.715	±0.0024	0.261	±0.0025
303.15	0.815	±0.0024	0.736	±0.0024	0.269	±0.0024
308.15	0.839	±0.0025	0.756	±0.0025	0.277	±0.0024
313.15	0.864	±0.0025	0.775	±0.0023	0.287	±0.0025
318.15	0.889	±0.0024	0.795	±0.0024	0.293	±0.0025
323.15	0.914	±0.0024	0.816	±0.0024	0.303	±0.0025
328.15	0.939	±0.0024	0.837	±0.0024	0.311	±0.0025
333.15	0.964	±0.0025	0.859	±0.0024	0.319	±0.0024
338.15	0.990	±0.0024	0.880	±0.0025	0.328	±0.0024
343.15	1.016	±0.0024	0.901	±0.0024	0.336	±0.0025

Table 3. Measured Mass Diffusion Coefficients D_{AB} of CH_4, O_2, CO_2 in Air from T = (278.15 to 343.15) K under Normal Atmospheric Pressure

	CH_4 (1) + air (2)		O_2 (1) + air (2)		CO_2 (1) + air (2)	
T	D_{AB}	$u_{D_{12}}$	D_{AB}	$u_{D_{12}}$	D_{AB}	$u_{D_{12}}$
K	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$	$cm^2 \cdot s^{-1}$
278.15	0.192	±0.0026	0.179	±0.0025	0.145	±0.0024
283.15	0.197	±0.0025	0.185	±0.0024	0.149	±0.0024
288.15	0.202	±0.0023	0.191	±0.0025	0.153	±0.0024
293.15	0.208	±0.0025	0.197	±0.0024	0.157	±0.0025
298.15	0.212	±0.0024	0.204	±0.0024	0.161	±0.0025
303.15	0.217	±0.0024	0.210	±0.0024	0.165	±0.0024
308.15	0.222	±0.0025	0.216	±0.0024	0.170	±0.0025
313.15	0.228	±0.0025	0.222	±0.0025	0.175	±0.0025
318.15	0.233	±0.0024	0.228	±0.0025	0.179	±0.0024
323.15	0.239	±0.0024	0.234	±0.0025	0.183	±0.0024
328.15	0.245	±0.0024	0.240	±0.0025	0.188	±0.0024
333.15	0.251	±0.0024	0.247	±0.0024	0.193	±0.0025
338.15	0.257	±0.0025	0.253	±0.0024	0.199	±0.0025
343.15	0.263	±0.0024	0.259	±0.0025	0.205	±0.0024

The comparison of the measured results and the calculated results is shown in Figure 4. The relative deviation is defined as

$$S_D = (D_i^{exp} - D_i^{cal})/D_i^{cal} \qquad (4)$$

where D_i^{cal} is the binary gas diffusion coefficient obtained with the correlation of Fuller and D_i^{exp} is the experimental binary gas diffusion coefficient. The average absolute deviation (S_{AAD}) is defined as

$$S_{AAD} = \frac{1}{N}\sum_{i=1}^{N} |(D_i^{exp} - D_i^{cal})/D_i^{cal}| \qquad (5)$$

From the comparison results, it can be seen that, in addition to the gas pair of O_2 (1) + air (2), the average absolute deviation is 9.0 %, and the average absolute deviation of the other five pairs of gas is within 5.0 %. Considering that the Fuller correlation has a uncertainty about 4.0 %,[13] the measured results in this work is reasonable.

For the binary diffusion coefficients of gases in air, the experimental data are scarce and only at around room temperature. In the present work, the experimental results in the H_2 (1)−air (2) system were compared with literature data[15] as shown in Figure 4. The results show that the average absolute deviation for the H_2 (1)−air (2) system between the measuring results in this work and the reference data is within 0.91 %.

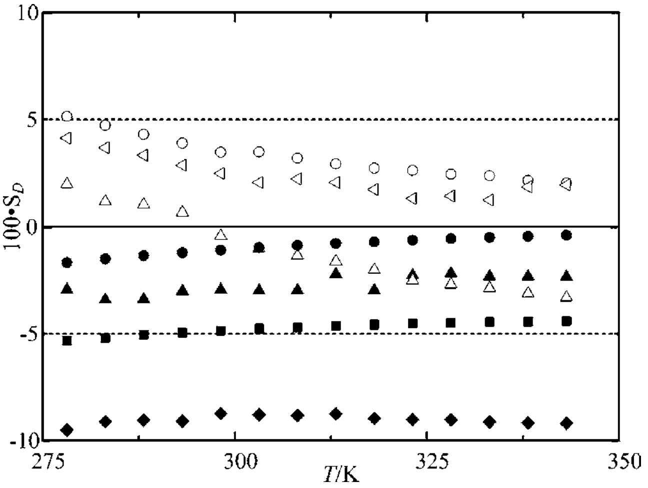

Figure 4. Relative deviation S_D obtained from eq 4 as a function of temperature T. ■, this work, representing the relative deviation of the binary gas diffusion coefficients of H_2 in air; ○, He; ▲, NH_3; △, CH_4; ◆, O_2; left-pointing triangle, CO_2; ●, literature data[15] of the binary gas diffusion coefficients of H_2 in air.

Conclusion

In this paper, on the basis of the Mach−Zehnder optical interference model, a digital real-time holographic interferometric experimental system was set up. A new diffusion cell especially for measuring the binary gas-phase diffusion coefficient was designed and constructed. Experimental theory for calculating the binary gas diffusion coefficient by object wave phase difference was introduced in detail. On this basis, the diffusion coefficients of H_2, He, NH_3, CH_4, O_2, and CO_2 in air at T = (278.15 to 343.15) K under atmospheric pressure were measured, and then the measuring results were compared with calculation results of the Fuller correlation and the literature data. The comparison results showed that measuring results is reasonable and credible and this laid a solid foundation for future measuring work.

Supporting Information Available:

Table A lists the measured binary diffusion coefficients of O_2 in air under standard conditions at different moments. Table B lists the physical properties of the gas studied in this work, as H_2, He, NH_3, CH_4, O_2, and CO_2. Tables C and D list the binary diffusion coefficients of H_2, He, NH_3, CH_4, O_2, and CO_2 in air calculated by the Fuller correlation and the relative deviation compared with the experimental results. Table E has a comparison of the measured binary diffusion coefficient of H_2 in air with the literature data. Figure A shows the optical system of Mach−Zehnder holographic interferometry. This material is available free of charge via the Internet at http://pubs.acs.org.

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