Mass Diffusion Coefficients of Oxygenated Fuel Additives in Diesel Oil and in Heptane at T = (276 to 333) K†

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Digital image holographic interferometry has been used to measure the mass diffusion coefficient of fluids. The mass diffusion coefficients of KCl in aqueous solution at a concentration pf 0.33 mol·dm^{-3} as the standard solution was used to verify the precision and reliability of the apparatus. The results show that the absolute average relative deviations is within 1.20 %. The uncertainty in mass diffusion coefficient is estimated to be not greater than ± 0.2 %. The mass diffusion coefficients of three different oxygenated fuel additives, bis(2-methoxyethyl)-ether (diethylene glycol dimethyl ether, DGM), 1,2-dimethoxyethane (ethyleneglycol dimethyl ether, GDME), and ethanol, were determined in both diesel oil and heptane at temperatures between (276 and 333) K.

Introduction

Diesel engines are used due to their relatively high fuel efficiency, but the exhaust emission contains considerable particulate matter (PM) and oxides of nitrogen (NO_x). Therefore, effort has been expended to reduce these emissions by the use of fuel additives. Oxygenated fuel additives are being used to reduce the effect of the emissions on global warming and the energy crisis.[1,2] In the past decade, there has been research in this field,[3,4] but there is little experimental data concerning the thermophysical properties of oxygenated fuel additives. The thermophysical properties are the basic parameters required to select substitute fuel and fuel additives. The mass diffusion coefficient is one of these properties and is related closely to the atomization and combustion process within combustion engines and is required for numerical simulation of the combustion process.[5] The prediction and measurement of mass diffusion coefficients are of increasing interest.

In this work, we modified an apparatus to measure the mass diffusion coefficient. To verify the operation, we determined the mass diffusion coefficients of KCl in aqueous solution at a concentration of 0.33 mol·dm^{-3} at T = 298.15 K. We also measured the mass diffusion coefficients of three different oxygenated fuel additives, bis(2-methoxyethyl)-ether (diethylene glycol dimethyl ether, DGM), 1,2-dimethoxyethane (ethyleneglycol dimethyl ether, GDME), and ethanol, in both diesel oil and heptane.

Experimental Section

Apparatus and Procedure. Measurements of the mass diffusion coefficient for oxygenated fuel additives were performed in a digital image holographic interferometer[6–9] shown in Figure 1. The light source was a LASER of wavelength 633 nm, with the beam expanded to a diameter of about 40 mm, and split by a prism into a reference beam and an object beam. The object beam passed through the diffusion cell into another prism to recombine it with the reference beam. Both the object and reference beams are projected onto the CCD camera, and holographic images obtained. Analysis of the interference fringes as a function of time permits the determination of mass diffusion coefficient. All the optical components are installed on an antivibration table to reduce the influence of the environmental shock.

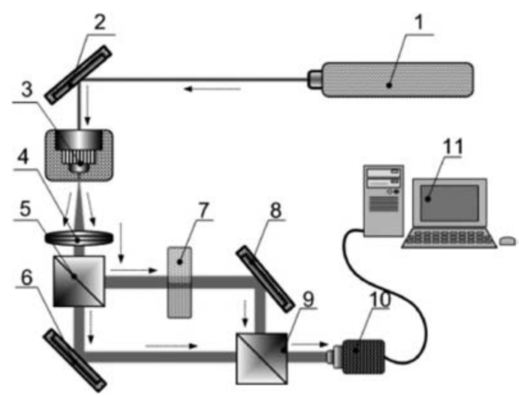

Figure 1. Optical system of digital image holographic interferometry. 1, He−Ne laser; 2, 6, 8, mirror; 3, spatial filter; 4, achromatic doublets lens; 5, 9, beam splitter prism; 7, diffusion cell; 10, CCD camera; 11, computer.

Our previous diffusion cell[10] has been modified as shown in Figure 2 by the addition of a sealed cavity between two optical surfaces to eliminate interference of the object beam channel and a variation of the optical surfaces to permit operation at higher pressures. The diffusion cell was thermostatted in a stirred fluid bath.

The fundamental of the mass diffusion coefficient measurement is based on Fick's second law for one-dimension diffusion,[11–14] of

$$\left(\frac{\partial c}{\partial t}\right) = D_{12}\left(\frac{\partial^2 c}{\partial z^2}\right) \tag{1}$$

where D_{12} is the mass diffusion coefficient; t is time; c is concentration; and z is the direction of diffusion.

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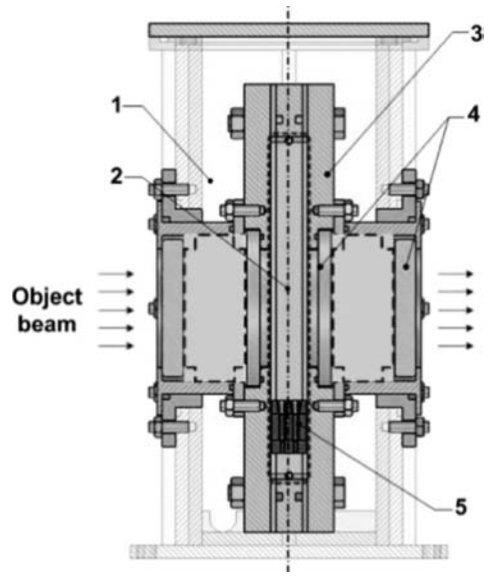

Figure 2. Diffusion system. 1, thermostatic water bath; 2, diffusion zone; 3, diffusion cell; 4, optical glass; 5, honeycomb.

From analysis of the linear relationship between the variations of the phase of the object beam with concentration, the D_{12} can be obtained from[10]

$$D_{12} = \Delta z_{\mathrm{m}}^2 \frac{t_1/t_2 - 1}{8t_1 \ln(t_1/t_2)} \tag{2}$$

where t_1 and t_2 are the times of getting two holograms and Δz_{m} is the distance between two extreme points of concentration changes, which is obtained from digital image processing. From eq 2, it can be seen that knowledge of Δz_{m} is key in the determination of mass diffusion coefficient.

The digital image processing is shown in Figure 3. The holograms obtained from the CCD were extracted from the noise using a wavelet method and then filtered into the frequency and traversed to the frequency domain within a rectangular window. The mean value of each row vector of the continual phase difference matrix, the line of continual phase difference, can be obtained. Finally, the value of Δz_{m} can be determined.

Test of the Experimental System. To verify the operation of the improved diffusion cell, the mass diffusion coefficient of KCl in aqueous solution at temperatures of 298.15 K and a concentration 0.33 $\mathrm{mol \cdot dm^{-3}}$ was measured. The holograph images from the experimental system and the digital image processing mentioned are show in Figure 4. Table 1 shows the values of the mass diffusion coefficients which were determined from comparing the hologram obtained at time t_1 with seven other different holograms determined at different times t_2. The average of the seven experimental results is $1.863 \cdot 10^{-5}\ \mathrm{cm^2 \cdot s^{-1}}$. Comparing the experimental results with the literature value,[15]

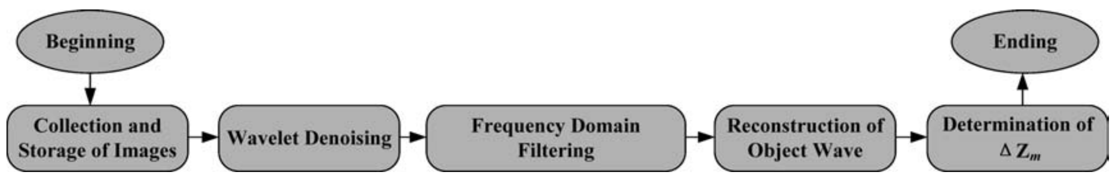

Figure 3. Digital image processing.

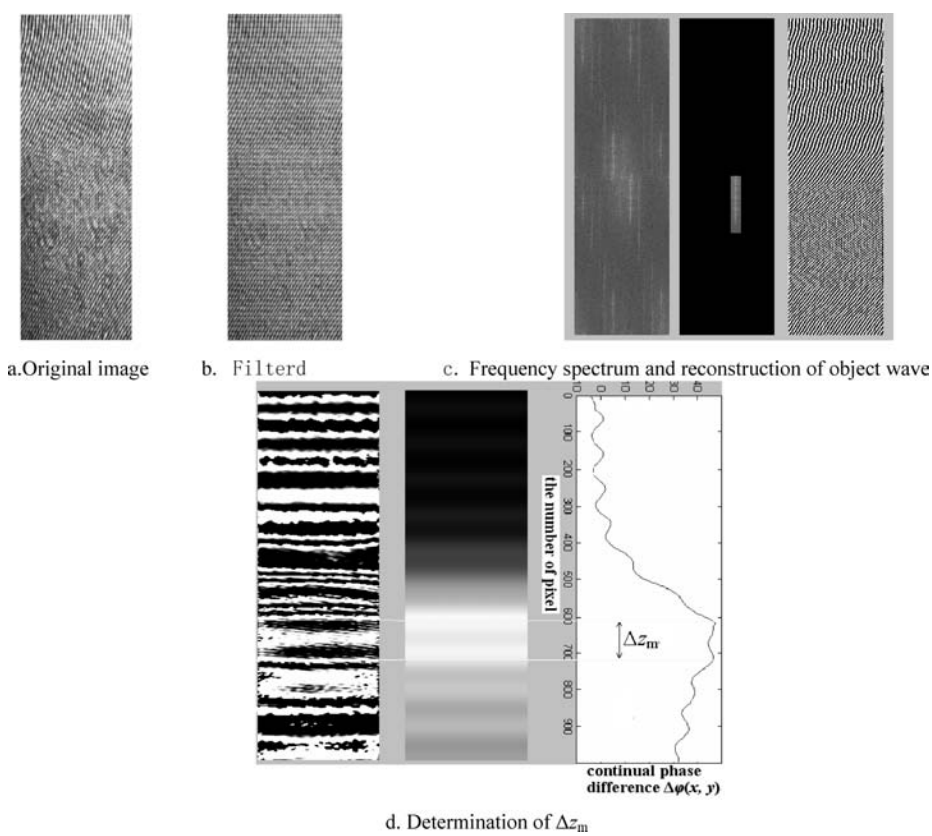

Figure 4. Digital image processing.

Table 1. Mass Diffusion Coefficients D_{12} and $\Delta D_{12} = (D_{12} - D_{ref})$ of KCl in Aqueous Solution at a Temperature of 298.15 K Obtained from Δz_m and Measurements at Time t_1 and t_2[a]

Δz_m/mm	t_1/s	t_2/s	$10^{-5}D_{12}$/cm^2·s^{-1}	$100 \cdot \Delta D_{12}/D_{ref}$
4.27	900	1800	1.826	−0.81
4.73	900	2700	1.884	2.34
4.93	900	3600	1.825	−0.87
5.23	900	4500	1.894	2.88
5.37	900	5400	1.865	1.30
5.47	900	6300	1.832	−0.49
5.83	900	8100	1.912	3.86
$10^{-5}D^*_{12}$/cm^2·s^{-1}			1.863	
$10^{-5}D_{ref}$/cm^2·s^{-1}			1.841	
$100 \cdot (D^*_{12} - D_{ref})/D_{ref}$			1.20	

[a] D_{12} is the experimental results; D^* is the average of experimental results; D_{ref} is the literature value. Uncertainties: $\delta(D_{12}) = \pm$ 0.2 %; $\delta(t_1) = \pm$ 0.002 s; $\delta(t_2) = \pm$ 0.002 s; $\delta(\Delta z_m) = \pm$ 0.1 %.

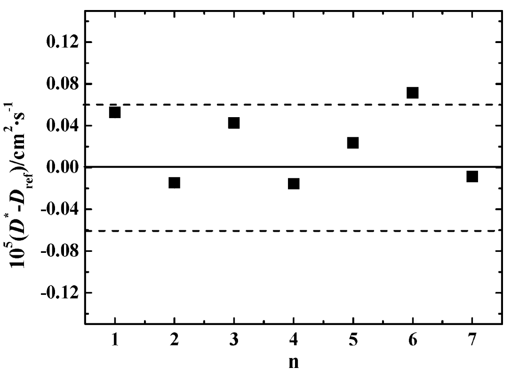

Figure 5. Absolute deviations of experimental results of KCl in aqueous solution. ■, experimental data.

Table 2. Sources of Uncertainty in the Measurement of Temperature and 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, $u_{c,T}$	0.08 K
mass diffusion coefficient	
time of getting hologram, u_5	0.001 %
distance between two extreme points of concentration, u_6	0.1 %
combined standard uncertainty $u_{c,D_{12}}$	0.1 %

Table 3. Molecular Formula, Molar Mass, Density, and Purity of Diesel, The Oxygenated Fuel Additives, Ethanol, and Heptane

	diesel oil	DGM	GDME	ethanol	heptane
molecular formula	$C_{10.8}H_{18.7}$	$C_6H_{14}O_3$	$C_4H_{10}O_2$	C_2H_6O	C_7H_{16}
molar mass/g·mol^{-1}	148.3	134.18	90.12	46.00	100.20
density/g·cm^{-3}	0.83	0.94	0.87	0.79	0.68
mass purity		≥ 99.5 %	≥ 98 %	≥ 99.7 %	≥ 97.0 %

Table 4. Composition of Diesel Oil

composition	alkane	cycloalkane	aromatics
mass percent	67.69 %	15.22 %	17.09 %

the maximum relative deviation is 3.86 % and the absolute average of relative deviations is within 1.20 %.

Figure 5 shows the absolute deviations of experimental results of KCl in aqueous solution at each experimental point. From Figure 4, it can be seen that the average deviations between experimental results and reference data[15] are about $\pm 0.06 \cdot 10^{-5}$ cm^2·s^{-1} (about 3 %).

Results and Discussion

Experimental Uncertainty. The mass diffusion coefficient determined from the holographic method is a function of temperature, the time at which the holograms were obtained, and the distance between two concentration extremes. To determine the uncertainty in the measurement, the uncertainty equation is used as follows.[16]

$$U_T = ku_{c,T} = k\sqrt{\Sigma(u_i)^2} \qquad (3)$$

$$U_{D_{12}} = ku_{c,D_{12}} = kD_{12}\left(\frac{u_{c,D_{12}}}{\bar{D}_{12}}\right) = kD_{12}\sqrt{\left(\frac{\partial \ln D_{12}}{\partial i}\right)^2 u_i^2} \qquad (4)$$

where U_T is the uncertainty in temperature; U_{D12} is the uncertainty of mass diffusion coefficient; i is the error source when the mass diffusion coefficient was measured; u_i is the uncertainty for each error source; and k is the coverage factor, which is taken as $k = 2$ with a confidence interval of 95 %. The uncertainties listed in Table 2 for temperature and mass diffusion coefficient were estimated to be not greater than $\pm$ 0.16 K and $\pm$ 0.2 %, respectively.

Table 5. Mass Diffusion Coefficients D_{12} Uncertainty u at Temperature T for GDME in Diesel Oil[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.91	3.819	± 0.007	304.35	33.47	± 0.07
278.36	4.559	± 0.009	307.60	45.57	± 0.11
281.49	6.194	± 0.002	311.06	56.20	± 0.11
283.39	7.303	± 0.002	312.93	68.83	± 0.13
286.99	8.408	± 0.031	315.68	72.08	± 0.17
288.77	9.139	± 0.019	318.53	90.06	± 0.20
291.08	9.631	± 0.022	322.39	103.1	± 0.2
293.91	12.57	± 0.03	326.88	128.6	± 0.3
295.69	15.89	± 0.04	328.41	158.7	± 0.3
297.82	17.33	± 0.04	331.89	173.1	± 0.4
301.54	22.11	± 0.05			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

Table 6. Mass Diffusion D_{12} Uncertainty u at Temperature T for Coefficients DGM in Diesel Oil[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.58	1.126	± 0.003	305.39	38.60	± 0.12
279.39	1.570	± 0.004	309.02	49.76	± 0.13
282.51	1.811	± 0.004	311.66	61.58	± 0.15
284.74	3.557	± 0.008	314.28	74.82	± 0.17
287.98	4.362	± 0.008	317.77	83.89	± 0.19
290.21	6.198	± 0.015	320.53	97.09	± 0.35
295.13	8.532	± 0.022	323.77	104.4	± 0.3
296.46	12.97	± 0.028	326.13	127.8	± 0.3
298.62	19.12	± 0.04	328.19	148.0	± 0.3
301.10	24.86	± 0.06	330.58	173.6	± 0.3
303.61	31.14	± 0.05			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

Table 7. Mass Diffusion Coefficients D_{12} Uncertainty u at Temperature T for Ethanol in Diesel Oil[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.32	1.736	± 0.004	306.29	14.65	± 0.03
279.87	2.027	± 0.005	308.40	20.59	± 0.05
282.11	2.664	± 0.006	311.21	25.94	± 0.07
284.39	3.019	± 0.006	314.45	27.59	± 0.07
288.52	3.541	± 0.007	318.82	36.26	± 0.12
291.79	4.392	± 0.010	320.43	41.77	± 0.09
293.72	5.274	± 0.012	322.77	52.16	± 0.14
296.83	6.697	± 0.017	326.91	57.55	± 0.12
299.64	8.768	± 0.018	329.98	68.97	± 0.17
300.49	10.05	± 0.026	332.36	76.91	± 0.16
303.16	12.82	± 0.027			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

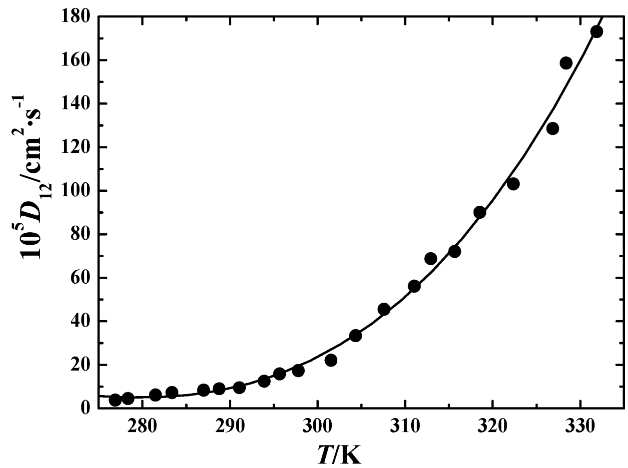

Figure 6. Mass diffusion coefficients of GDME in diesel oil. ●, experimental data; —, polynomial fit of experimental data.

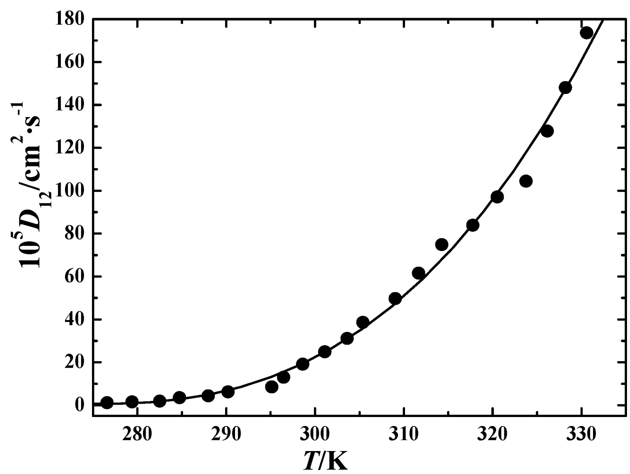

Figure 7. Mass diffusion coefficients of DGM in diesel oil. ●, experimental data; —, polynomial fit of experimental data.

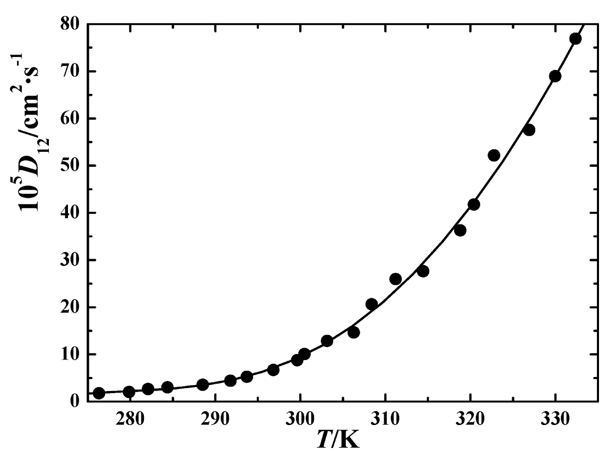

Figure 8. Mass diffusion coefficients of ethanol in diesel oil. ●, experimental data; —, polynomial fit of experimental data.

Experimental Results. In this work, the mass diffusion coefficients of three different oxygenated fuel additives, bis(2-methoxyethyl)-ether, 1,2-dimethoxyethane, and ethanol in both diesel oil and heptane were determined at 21 temperatures between (276 and 333) K. Heptane was used in this work as it is considered a reference fluid for diesel oil because of the closed cetane number between heptane and diesel oil. The chemical formula and molar mass of the materials used in this work are listed in Table 3, while the chemical composition of diesel oil is listed in Table 4.

Table 8. Mass Diffusion Coefficients D_{12} Uncertainty u at Temperature T for GDME in Heptane[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.64	5.243	± 0.012	304.98	5.886	± 0.013
277.73	5.275	± 0.016	306.86	5.982	± 0.016
281.59	5.313	± 0.011	310.67	6.129	± 0.010
284.32	5.351	± 0.013	312.41	6.296	± 0.014
287.28	5.396	± 0.013	316.09	6.478	± 0.013
289.59	5.437	± 0.012	319.27	6.619	± 0.015
292.83	5.513	± 0.012	322.81	6.786	± 0.020
294.95	5.567	± 0.016	325.36	6.939	± 0.015
296.37	5.634	± 0.014	328.78	7.295	± 0.016
298.46	5.692	± 0.010	332.63	7.593	± 0.015
301.29	5.793	± 0.012			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

Table 9. Mass Diffusion Coefficients D_{12} Uncertainty u at Temperature T for DGM in Heptane[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.64	0.942	± 0.0028	304.98	1.116	± 0.0020
277.73	0.948	± 0.0017	306.86	1.141	± 0.0030
281.59	0.951	± 0.0022	310.67	1.178	± 0.0021
284.32	0.958	± 0.0021	312.41	1.225	± 0.0027
287.28	0.976	± 0.0022	316.09	1.258	± 0.0025
289.59	0.987	± 0.0025	319.27	1.314	± 0.0031
292.83	1.005	± 0.0022	322.81	1.369	± 0.0033
294.95	1.024	± 0.0022	325.36	1.428	± 0.0029
296.37	1.046	± 0.0023	328.78	1.484	± 0.0041
298.46	1.068	± 0.0024	332.63	1.529	± 0.0033
301.29	1.084	± 0.0020			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

Table 10. Mass Diffusion Coefficients D_{12} Uncertainty u at Temperature T for Ethanol in Heptane[a]

T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}	T/K	10^5D_{12}/cm^2·s^{-1}	$10^5u_{D_{12}}$/cm^2·s^{-1}
276.64	7.572	± 0.015	304.98	8.224	± 0.018
277.73	7.613	± 0.021	306.86	8.365	± 0.020
281.59	7.641	± 0.015	310.67	8.471	± 0.018
284.32	7.695	± 0.017	312.41	8.582	± 0.018
287.28	7.754	± 0.014	316.09	8.712	± 0.019
289.59	7.786	± 0.017	319.27	8.906	± 0.018
292.83	7.841	± 0.016	322.81	9.174	± 0.029
294.95	7.898	± 0.013	325.36	9.219	± 0.020
296.37	7.936	± 0.020	328.78	9.361	± 0.024
298.46	8.067	± 0.020	332.63	9.503	± 0.019
301.29	8.193	± 0.015			

[a] Uncertainties: $\delta(T) = \pm$ 0.16 K.

The mass diffusion coefficients of the oxygenated fuel additives in diesel oil at temperatures between (276 and 333) K at an atmospheric pressure of 0.1 MPa are listed in Tables 5, 6, and 7 and shown in Figures 6, 7, and 8. The estimated experimental uncertainty, obtained from Table 2, at each state points (as $u_{D_{12}}$) are also listed in Tables 5, 6, and 7.

The measured mass diffusion coefficients of the oxygenated fuel additives with heptane at temperatures of (276 to 333) K are listed in Tables 8, 9, and 10, with the estimated experimental uncertainty, and shown in Figures 9, 10, and 11.

According to the experiment results, the mass diffusion coefficients of DGME (1), DGM (2), and ethanol (3) in diesel oil (4) were fit to the following equations, respectively:

$$D_{14}/\mathrm{cm^2\cdot s^{-1}} = -9238.425 + 108.567(T/\mathrm{K}) - 0.422(T/\mathrm{K})^2 + 5.430\cdot 10^{-4}(T/\mathrm{K})^3 \quad (5)$$

$$D_{24}/\text{cm}^2\cdot\text{s}^{-1} = -9791.273 + 113.421(T/\text{K}) - 0.436(T/\text{K})^2 + 5.572\cdot10^{-4}(T/\text{K})^3 \quad (6)$$

and

$$D_{34}/\text{cm}^2\cdot\text{s}^{-1} = -3824.600 + 45.469(T/\text{K}) - 0.178(T/\text{K})^2 + 2.310\cdot10^{-4}(T/\text{K})^3 \quad (7)$$

The mass diffusion coefficients of DGME (1), DGM (2), and ethanol (3) in heptane (5) were fit to the following equations, respectively

$$D_{15}/\text{cm}^2\cdot\text{s}^{-1} = -50.310 + 0.676(T/\text{K}) - 0.275\cdot10^{-2}(T/\text{K})^2 \quad (8)$$

$$D_{25}/\text{cm}^2\cdot\text{s}^{-1} = 12.642 - 0.086(T/\text{K}) + 1.598\cdot10^{-4}(T/\text{K})^2 \quad (9)$$

and

$$D_{35}/\text{cm}^2\cdot\text{s}^{-1} = 35.156 - 0.212(T/\text{K}) + 4.091\cdot10^{-4}(T/\text{K})^2 \quad (10)$$

For the liquid diffusion, the Stokes−Einstein equation

$$D_{12} = \frac{RT}{6\pi\eta_B r_A} \quad (11)$$

was used to determine the correlation between mass diffusion coefficient and viscosity of solvent. In eq 11, η_B is the solvent viscosity, and r_A is the radius of the solute sphere. Equation 11 shows that the mass of the diffusion coefficient is inversely proportional to r_A. For the oxygenated fuels used in our experiment, the radius of the solute sphere decreases in the order r_A(DGM) > r_A(GDME) > r_A(ethanol). According to eq 11 the mass diffusion coefficient should follow the order D_{12}(DGM) > D_{12}(GDME) > D_{12}(ethanol). In our work, the value of mass diffusion coefficient in heptane, which increases from ethanol, to GDME, and to DGM, obeys the form anticipated by eq 11. However, for diesel oil, the value of mass diffusion coefficient was found to increase with solvent in the order ethanol, GDME, and DGM, which is different from the order expected from viscosity and eq 11.

Conclusions

In this work, a modified digital image holographic interferometry diffusion cell was used to measure the mass diffusion coefficient. The operation of the apparatus was determined with measurements of the mass diffusion coefficients of KCl in aqueous solution at concentration of 0.33 mol·dm^{-3} and comparison with the literature values that showed the absolute average of relative deviation is within 1.20 %.

The apparatus was used to measure the mass diffusion coefficients of three different oxygenated fuel additives, bis(2-methoxyethyl)-ether, 1,2-dimethoxyethane, and ethanol, in both diesel oil and heptane at temperatures between (276 and 333) K. The experimental uncertainties in temperature and in mass diffusion coefficient are estimated to be not greater than ± 0.16 K and ± 0.2 %, respectively.

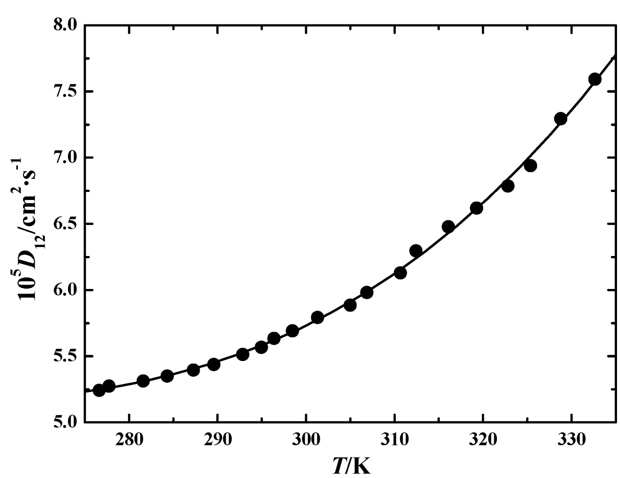

Figure 9. Mass diffusion coefficients of GDME in heptane. ●, experimental data; —, polynomial fit of experimental data.

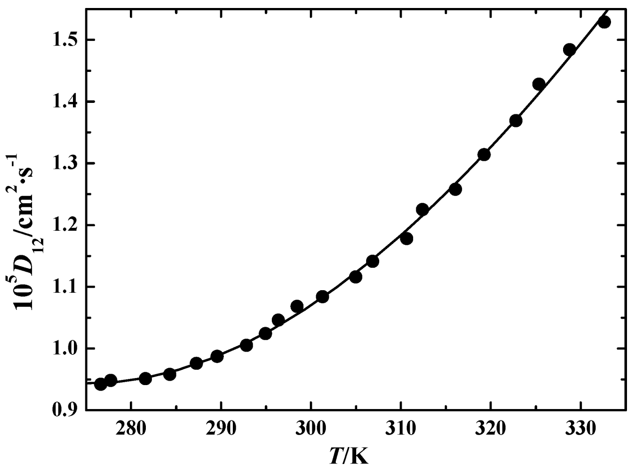

Figure 10. Mass diffusion coefficients of DGM in heptane. ●, experimental data; —, polynomial fit of experimental data.

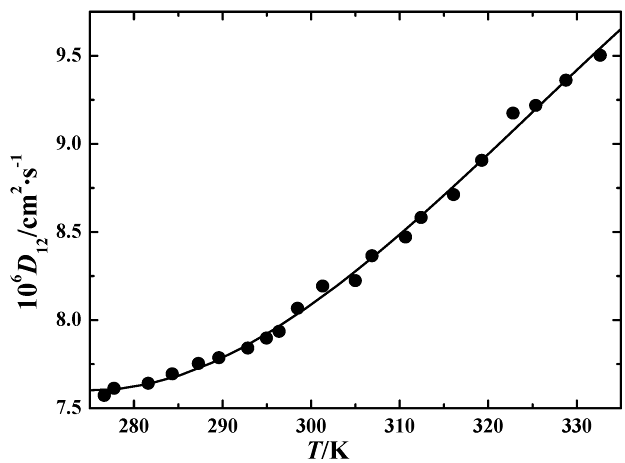

Figure 11. Mass diffusion coefficients of ethanol in heptane. ●, experimental data; —, polynomial fit of experimental data.

Supporting Information Available:

Table A. Properties of DGM, GDME, and Ethanol. Table B. Properties of Heptane. This material is available free of charge via the Internet at http://pubs.acs.org.

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