J. Chem. Eng. Data **1994,** *39,* 911–915

Binary Diffusion Coefficients of Organic Compounds in Hexane, Dodecane, and Cyclohexane at 303.2–333.2 K and 16.0 MPa

Toshitaka Funazukuri,* Norihiro Nishimoto, and Noriaki Wakao

Department of Materials Science and Chemical Engineering, Yokohama National University, Yokohama 240, Japan

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Binary diffusion coefficients for infinitely dilute solutions of acetone, benzene, toluene, *m*-xylene, 1,3,5-trimethylbenzene, and naphthalene in hexane, in dodecane, and in cyclohexane were measured from 303.2 to 333.2 K and at 16.0 MPa using the Taylor dispersion technique. In addition, the measurements for *o*- and *p*-xylenes, vitamin K_3, linoleic acid methyl ester, indole, *dl*-limonene, vitamin A acetate, vitamin E, and vitamin K_1 in hexane and vitamin K_3 and linoleic acid methyl ester in dodecane were made at the limited conditions. The D_{12} results in the three solvents are expressed by the equation $D_{12}/T = \alpha\eta^{\beta}$ with average absolute deviation of 5.2%, where η is the solvent viscosity and α and β are constants related to the solute molar volume at the normal boiling point. For the D_{12} measured in this study this correlation is as accurate as Nakanishi's correlation which is superior to other correlations examined.

Introduction

Binary diffusion coefficients D_{12} of organic compounds in liquid solvents are of importance not only in chemical engineering design but also in the theory of transport properties. Fewer measurements of D_{12}, in particular at high pressures, have been made, as compared with the other transport properties such as viscosity and thermal conductivity. The Taylor dispersion technique (*1, 2*) has been commonly used to measure D_{12} in gaseous, liquid, and supercritical fluids, as reviewed in refs 3–6 because of relatively high accuracy (~1%) and the short amount of time required (*4*). We measured binary diffusion coefficients of some unsaturated fatty acid derivatives and vitamins in supercritical carbon dioxide at 16.0 MPa (*7–9*). In this study, binary diffusion coefficients for infinitely dilute solutions of various organic compounds in hexane, in dodecane, and in cyclohexane were measured mainly at 16.0 MPa by the Taylor dispersion technique, and the measured D_{12} values were correlated with the solvent viscosity and temperature.

Theory

The theoretical basis for measuring binary diffusion coefficients in a cylindrical tube was developed by Taylor (*1*) and Aris (*2*). Alizadeh et al. (*10*) analyzed rigorously the fundamental equation of the Taylor dispersion by the moment method, and evaluated the corrections of a practical instrument from the ideal. The theoretical background and the calculation procedures of the D_{12} determination were described elsewhere (*7–9, 11*).

The concentration vs time curve obtained experimentally is compared in the time domain with the curves calculated with assumed values of the apparent binary diffusion coefficients D'_{12}. The value of D'_{12} is then determined so that error,

$$\text{error} = (\Sigma\{C^{+}_{\text{exp}}(t_i) - C^{+}_{\text{cal}}(t_i)\}^2\Delta t/\Sigma\{C^{+}_{\text{exp}}(t_i)\}^2\Delta t)^{1/2} \quad (1)$$

is minimized (error < 0.013). This determined value of D'_{12} is the binary diffusion coefficient D_{12} at its infinite dilution unless the D'_{12} value is affected by the solvent flow rate u.

Experiments

Table 1 lists the molecular mass M, molar volume V_b at its normal boiling point, estimated by the method of Le Bas (*12*), and purity of the solutes and the solvents used in this study. These reagents were used without further purification.

The experimental apparatus, designed for the measurements at elevated pressures, is substantially identical to that used in our previous studies (*7–9, 11*). The pump assembly and the back-pressure regulator were replaced.

A microfeeder pump (model CAP-L02, JEOL Moleh, Japan) was used to supply the solvents hexane, dodecane, and cyclohexane. This pump, which is capable of feeding the solvents at constant flow rates higher than 0.1 μL/min with ±0.1% precision, consists of two plunger pumps connected in cascade which can fully compensate volume loss due to compression with an intelligent controller.

The pressure of the solvent flow was regulated with an accuracy of ±0.1 MPa by the back-pressure regulator (model CAP BP01, JEOL Moleh), equipped with a temperature controller and having a small dead volume (~5 μL). The pressures in the diffusion column were continuously monitored with pressure sensors both at the pump head and just upstream of the back-pressure regulator. The pressure gauge was also installed just upstream of the injector. The pressure drop between the injector and the back-pressure regulator was within 0.1 MPa during the measurements. Almost no pressure pulsation was observed.

Results and Discussion

Measured D_{12} Values. Measured binary diffusion coefficients in hexane, in dodecane, and in cyclohexane are presented in Tables 2–4, respectively. Densities and viscosities of the solvents were estimated from the literature data for hexane (*13–16*), dodecane (*17, 18*), and cyclohexane (*18–20*).

A solute (diluted at a concentration of ~2–30 wt % with the same solvent as that flowing in the diffusion column) as a tracer species was injected through a 0.5-μL sample loop. It was found for benzene solute that there was scarcely a difference between the injection of the neat solute and that of the diluted one (~10–50 vol %) in the measured

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* To whom correspondence should be addressed.

0021-9568/94/1739-0911$04.50/0

Table 1. Solutes and Solvents Used in This Study

compound		M	V_b (cm^3·mol^{-1})	purity %	supplier
		Solute			
acetone	C_3H_6O	58.08	74.0	99.5	Tokyo Kasei
benzene	C_6H_6	78.11	96.0	99.5	Junsei
toluene	C_7H_8	92.13	118	99	Tokyo Kasei
m-xylene	C_8H_{10}	106.2	140	98	Tokyo Kasei
indole	C_8H_7N	117.1	130	99	Aldrich
1,3,5-trimethylbenzene	C_9H_{12}	120.2	163	97	Tokyo Kasei
naphthalene	$C_{10}H_8$	128.2	148	99	Aldrich
dl-limonene	$C_{10}H_{16}$	136.2	192	~96	Wako
cis-Jasmone	$C_{11}H_{16}O$	164.2	218	90	Aldrich
vitamin K$_3$	$C_{11}H_8O_2$	172.2	177	98.5	Wako
linoleic acid methyl ester	$C_{19}H_{34}O_2$	294.5	425	99	Sigma
vitamin A acetate	$C_{22}H_{32}O_2$	328.5	449	~90	Sigma
vitamin E	$C_{29}H_{50}O_2$	430.7	603	98	Tokyo Kasei
vitamin K$_1$	$C_{31}H_{46}O_2$	450.7	614	97	Wako
		Solvent			
hexane	C_6H_{14}	86.17	141	[a]	Junsei
dodecane	$C_{12}H_{26}$	170.3	274	99.6	Japan Energy
cyclohexane	C_6H_{12}	84.16	118	[a]	Junsei

[a] Spectroscopic grade.

Table 2. Measured Binary Diffusion Coefficients in Hexane

solute	λ^a/nm	$10^9 D_{12}$/(m^2·s^{-1})				
	T/K	303.2	313.2	313.2	323.2	333.2
	P/MPa	16.0	16.0	25.0	16.0	16.0
	ϱ/(kg·m^{-3})	666	658	667	650	641
	η/(mPa·s)	0.332	0.304	0.329	0.279	0.257
acetone	280	4.46	5.29	4.95	5.88	6.23
benzene	260	3.89[b]	4.48[b]	3.91	4.86[b]	5.27[b]
toluene	265	3.79[b]	4.32[b]	3.82	4.76[b]	5.24[b]
m-xylene	270	3.44[b]	3.63	3.40	4.37[b]	4.78[b]
o-xylene	280		3.74	3.32		
p-xylene	280		3.69	3.41		
1,3,5-trimethylbenzene	270	3.10[b]	3.55[b]	3.11	3.91[b]	4.26[b]
naphthalene	275−290	3.54[b]	3.70[b]	3.32	4.33[b]	5.03[b]
vitamin K$_3$	250		3.12[b]	3.07		
linoleic acid methyl ester	205		1.90[b]	1.82		
indole	265		3.78[b]	3.47		
dl−limonene	295		3.12			
vitamin A acetate	340		1.92			
vitamin E	305		1.52			
vitamin K$_1$	335		1.55			

[a] Wavelength for measuring D_{12}. [b] Reported in ref 11.

Table 3. Measured Binary Diffusion Coefficients in Dodecane

solute	λ^a/nm	$10^9 D_{12}$/(m^2·s^{-1})				
	T/K	303.2	313.2	323.2	333.2	343.2
	P/MPa	16.0	16.0	16.0	16.0	16.0
	ϱ/(kg·m^{-3})	753	746	739	733	726
	η/(mPa·s)	1.51	1.28	1.10	0.961	0.849
acetone	280	1.58	1.81	2.20	2.57	2.92
benzene	260	1.36	1.61[b]	1.87	2.20	
toluene	260	1.28	1.49[b]	1.74	2.07	
m-xylene	260−265	1.16	1.35[b]	1.57	1.84	
1,3,5-trimethylbenzene	265	0.974	1.19[b]	1.36	1.55	
naphthalene	285	1.07	1.26[b]	1.52	1.73	1.90
vitamin K$_3$	250	0.911	1.11[b]	1.29	1.49	
linoleic acid methyl ester	210	0.442	0.552	0.661	0.789	

[a] Wavelength for measuring D_{12}. [b] Reported in ref 11.

values of binary diffusion coefficients within the experimental errors.

To confirm no effect of the secondary flow on D_{12}, the measurements were made 5−10 times for every measurement condition of each solute by changing the flow rates u in the range from 3×10^{-3} to 8×10^{-3} m/s. Figure 1 shows plots of the apparent binary diffusion coefficient D'_{12} of linoleic acid methyl ester at 313.2 K and 16.0 MPa vs hexane flow rate u, together with the corresponding De^{2}Sc value. Almost no secondary flow effect on D'_{12} was observed in the u range studied.

Lauer et al. (*21*) also mentioned that the effect of secondary flow on D_{12} was negligible if eq 2 was satisfied when K was equal to 100. De is the Dean number, defined

$$\mathrm{De}^2\mathrm{Sc} < K \qquad (2)$$

as $(2\varrho u a_0/\eta)(a_0/R_c)^{1/2}$, where ϱ is the solvent density, a_0 is

Table 4. Measured Binary Diffusion Coefficients in Cyclohexane

solute	λ^a/nm	$10^9D_{12}/(\mathrm{m^2 \cdot s^{-1}})$			
	T/K	303.2	313.2	323.2	333.2
	P/MPa	16.0	16.0	16.0	16.0
	$\varrho/(\mathrm{kg \cdot m^{-3}})$	782	774	765	756
	$\eta/(\mathrm{mPa \cdot s})$	1.00	0.853	0.742	0.656
acetone	280	1.96	2.31	2.71	3.15
benzene	255	1.54	1.91	2.16	2.51
toluene	260	1.57	1.81	2.10	2.42
m-xylene	265	1.39	1.69	1.88	2.17
1,3,5-trimethylbenzene	265	1.23	1.45	1.65	1.93
naphthalene	275	1.31	1.52	1.79	2.06
vitamin K_3	250		1.38^b		
linoleic acid methyl ester	210		0.780^b		
indole	265		1.63^b		
cis-jasmone	230		1.09^b		

^{a} Wavelength for measuring D_{12}. ^{b} Reported in ref 11.

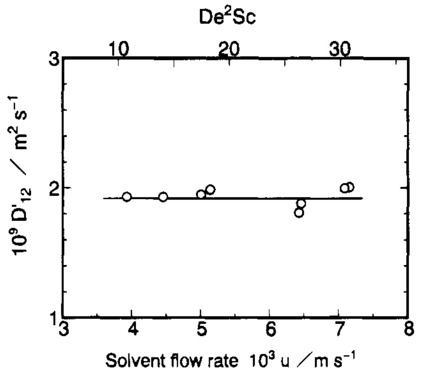

Figure 1. Apparent diffusion coefficient D'_{12} vs solvent flow rate u for linoleic acid methyl ester in hexane at 313.2 K and 16.0 MPa, together with the corresponding De^{2}Sc value.

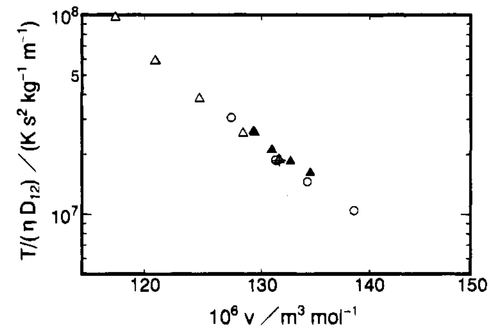

Figure 2. $T/\eta D_{12}$ vs hexane volume v for measured D_{12} values (▲) of benzene, as compared with the literature values: (+) Shankland et al. (*22*); (○) Dymond (*23*); (△) Dymond and Woolf (*24*).

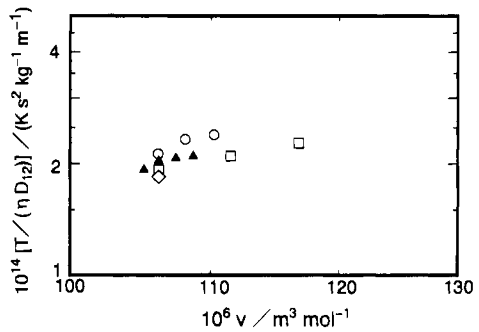

Figure 3. $T/\eta D_{12}$ vs cyclohexane molar volume v for measured D_{12} values (▲) of toluene, as compared with the literature values: (◇) Holmes et al. (*25*); (○) Sanni and Hutchison (*26*); (□) Sun and Chen (*27*).

the tubing radius, η is the solvent viscosity, and R_c is the coil radius, and Sc is the Schmidt number ($=\eta/\varrho D_{12}$). Alizadeh et al. (*10*) estimated the error to be ~0.05% when the K value was equal to 20. In this study the De^{2}Sc values for most measurements are less than 50, which corresponds to ~0.5% deviation (*10*).

In the present measurements the reproducibility of the mean residence times of the response curves is within 1%, and the fitting error is less than 1.3%, as mentioned above. The deviation due to the secondary flow is 0.5%. Consequently, the accuracy in the present measurements is estimated to be 3%.

Comparison of the Measured D_{12} Values with the Literature Data. Figure 2 shows the comparison of the present D_{12} data of benzene in hexane at 303.2–333.2 K and 16.0 MPa with those reported in the literature (*22–24*). Shankland et al. (*22*) used the Gouy technique to measure D_{12} at 298 K and atmospheric pressure. Dymond (*23*) measured D_{12} at 273.2–333.2 K and atmospheric pressure using both the diaphragm cell and the Taylor dispersion methods. Dymond and Woolf (*24*) measured D_{12} at 298.2 K and 0.1–400 MPa with the diaphragm cell method. Since these measurement conditions are different, the D_{12} values cannot be compared directly. Thus, the D_{12} data are compared in plots of $T/(\eta D_{12})$ vs solvent molar volume v. Note that the values of the solvent densities and viscosities were estimated, while these are presented in some papers. The values of $T/\eta D_{12}$ decrease with decreasing hexane molar volume v, as shown in this figure, and the present data are consistent with the literature data (maximum difference of 10%). The present D_{12} data for toluene, naphthalene, and 1,3,5-trimethylbenzene in hexane are also consistent with the literature values (not shown in the figures). The maximum deviations from the literature values in plots of $T/\eta D_{12}$ vs v are 5% for toluene (*22–24*), 5% for naphthalene (*23*), and 3% for 1,3,5-trimethylbenzene (*23*).

Figure 3 shows the same plots for D_{12} of toluene in cyclohexane. The D_{12} data of Holmes (*25*) and Sanni and Hutchison (*26*) were measured at atmospheric pressure with the diaphragm cell method. Sun and Chen (*27*) measured D_{12} at elevated pressures with the Taylor dispersion method. The values of $T/\eta D_{12}$ slightly increase with increasing cyclohexane molar volume. The difference between the present data and the literature values (*25, 27*) is within 6%.

Correlation with the Solvent Viscosity. It is known (*6, 28–33*) that binary diffusion coefficients can be correlated with the solvent viscosities, as expressed by eq 3.

$$D_{12}/T = \alpha\, \eta^{\beta} \tag{3}$$

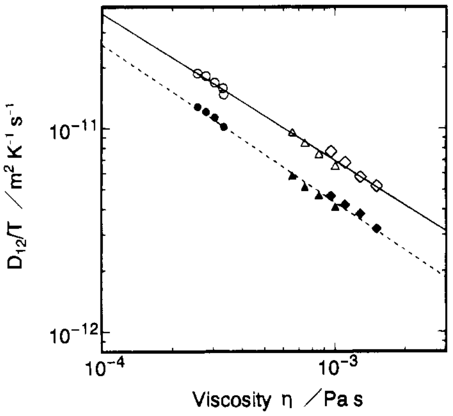

Figure 4. D_{12}/T vs solvent viscosity η at 16.0 MPa and 303.2–333.2 K for D_{12} of acetone in hexane (○), cyclohexane (△), and dodecane (◇), and for D_{12} of 1,3,5-trimethylbenzene in hexane (●), cyclohexane (▲), and dodecane (◆).

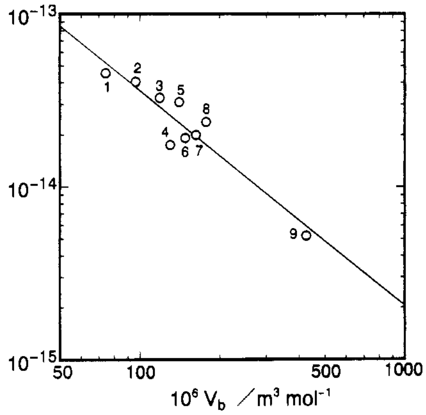

Figure 5. α vs solute molar volume V_b at its normal boiling point for each solute measured in this study (1, acetone; 2, benzene; 3, toluene; 4, indole; 5, *m*-xylene; 6, naphthalene; 7, 1,3,5-trimethylbenzene; 8, vitamin K_3; 9, linoleic acid methyl ester).

α and β are constants, and the units of D_{12}, T, and η are m^2 s^{-1}, K, and Pa s, respectively. Equation 3 is valid for D_{12} data of acetone and 1,3,5-trimethylbenzene in the three solvents, as shown in Figure 4. The plots are found to be expressed by a single straight line for each solute. Thus, both constants are correlated with a solute physical property, or solute molar volume V_b at its normal boiling point, as shown in Figures 5 and 6. From both plots, the values of α and β in eq 3 are

$$\alpha = 3.718 \times 10^{-19} V_b^{-1.246} \qquad (4a)$$

and

$$\beta = -1.981 V_b^{0.1072} \qquad (4b)$$

where the unit of V_b is m^3 mol^{-1}, and the values are listed in Table 1. The average absolute deviation, AAD (defined by eq 5), for eq 3 with the values of α and β calculated by eqs 4a and 4b, respectively, is 5.2% with the number of data points ND = 106 for the three solvents.

$$\mathrm{AAD}/\% = \frac{100}{\mathrm{ND}}\sum_{1}^{\mathrm{ND}}\left|\frac{D_{12,\mathrm{pred}}}{D_{12,\mathrm{exp}}} - 1\right| \qquad (5)$$

Figure 7 shows the deviations of the D_{12} values predicted by eq 3 from those measured for the three solvents. The deviations for most D_{12} values are within ±10%.

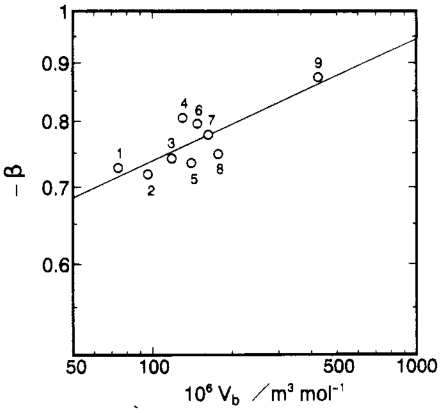

Figure 6. $-\beta$ vs solute molar volume V_b at its normal boiling point for each solute measured in this study (the numbers designated are the same as in Figure 5).

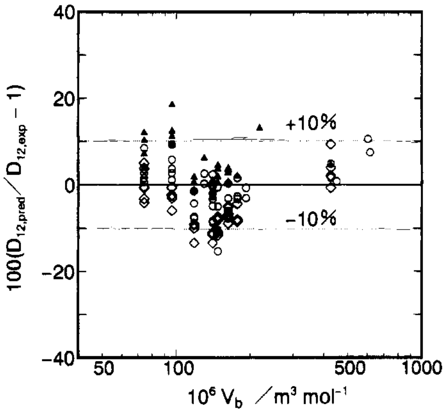

Figure 7. Accuracy in the correlation (eq 3) for the three solvents hexane (○), cyclohexane (▲), and dodecane (◇).

Table 5. Accuracy in the Empirical Correlations Examined for D_{12} Data Listed in Tables 2–4

	AAD[a]/%			
correlation	hexane ND = 44	dodecane ND = 34	cyclohexane ND = 28	all ND = 106
present study (eq 3)	4.3	6.6	5.1	5.2
Wilke–Chang (*12*)	3.9	6.5	18.0	8.5
Scheibel (*34*)	36.6	31.7	8.7	27.7
Reddy–Doraiswamy (*35*)	11.2	36.7	20.0	21.7
Lusis–Ratcliff (*36*)	11.2	7.0	11.3	9.9
Nakanishi (*37*)[b]	6.2	6.2	5.7	6.1
Hayduk–Minhas (*38*)[c]	17.1	23.0		19.7[d]
Chen–Chen (*39*)	6.6	16.0	20.5	13.3
Wong–Hayduk (*40*)[c]	13.1	18.5		15.5[d]

[a] AAD = average absolute deviation defined by eq 5. [b] Modified by Reid et al. (*41*). [c] Correlation for *n*-paraffin solvents. [d] AAD for D_{12} in hexane and dodecane.

Examination of the Empirical Correlations. The accuracy in the various empirical correlations (*12*, *34–40*) is examined with the D_{12} measured in this study. Table 5 lists the AAD values. Nakanishi's correlation (*37*) is found to be the most accurate (AAD = 6.1%) among the empirical correlations examined, but eq 3 is slightly better. Note that Nakanishi originally proposed the correlation for D_{12} at 298.2 K, and Reid et al. (*41*) extended it to the prediction at different temperatures by assuming $T/\eta D_{12}$ to be constant.

Figure 8 shows plots of D_{12} measured vs solute molar volume V_b at its normal boiling point, estimated by the method of Le Bas (*12*), for the three solvents at 313.2 K and 16.0 MPa. The three slopes are nearly 0.6 which is consistent with that in the Wilke–Chang equation (*12*). However, the use of the Wilke–Chang equation is limited

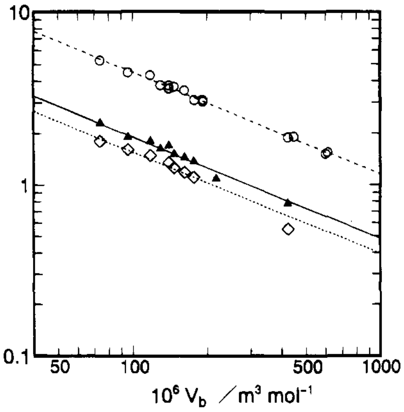

Figure 8. Binary diffusion coefficient D_{12} at 313.2 K and 16.0 MPa vs solute molar volume V_b at its normal boiling point for the three solvents hexane (○), cyclohexane (▲), and dodecane (◇).

at higher pressures because the values of $T/\eta D_{12}$ indicate the pressure dependence as shown in Figures 2 and 3. The AAD value for the Wilke–Chang equation is 8.5% for the three solvents, and the accuracy is poor for the cyclohexane solvent. The other correlations except for Nakanishi's are less accurate as listed in Table 5.

Conclusions

Binary diffusion coefficients for infinitely dilute solutions of some organic compounds in hexane, in dodecane, and in cyclohexane were measured mainly at 303.2–333.2 K and 16.0 MPa by the Taylor dispersion method. The data of D_{12} measured were expressed by eq 3 (accuracy of 5.2%), and the two constants involved in eq 3 were correlated with solute molar volume at normal boiling point. The present correlation was as good as Nakanishi's correlation which is the most accurate among other empirical equations examined.

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Received for review May 2, 1994. Accepted July 29, 1994.⊗ The authors are grateful to the Ministry of Education of Japan for financial support in the form of a grant-in-aid for a priority area.

⊗ Abstract published in *Advance ACS Abstracts,* September 1, 1994.