

face tension for the ternary melts. Judging from the shape of the contour, the lowest bottom of the surface tension isotherm seems to be near the point of $\text{LiF}:\text{BeF}_2 = 2:1$ on the $\text{LiF}-\text{BeF}_2$ axis, and the formation of BeF_4^{2-} is almost essentially complete in the ternary system as well as in the $\text{LiF}-\text{BeF}_2$ binary melts.

Conclusion

The surface tensions of $\text{LiF}-\text{ThF}_4$ binary melts and $\text{LiF}-\text{BeF}_2-\text{ThF}_4$ ternary melts were measured by the maximum bubble pressure method. The surface tension isotherm for the binary melts showed a slight negative deviation from additivity and a minimum near the composition of $\text{LiF}:\text{ThF}_4 = 2:1$. The addition of BeF_2 into the $\text{LiF}-\text{ThF}_4$ binary melts brought an abrupt decrease of the surface tension of the ternary melts. The former result may be attributed to a partial formation of a complex ion such as ThF_6^{2-} and the latter seems to be due to a complete formation of BeF_4^{2-} in the ternary melts.

These data are expected to be useful in handling the fuel solvent of MSBR.

Registry No. LiF , 7789-24-4; ThF_4 , 13709-59-6; BeF_2 , 7787-49-7.

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Diffusion of Propane in Helium

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The apparatus used by Yang and Hawkes had been modified to give greater mechanical reliability and its reliability has been demonstrated. The diffusion coefficients of a *n*-propane in helium at 1.0 atm from 21.3 to 60.3 °C were determined by using the new apparatus and follow the equation $\log D_{12} = -8.1277 + 3.0805 \log T$ with a correlation coefficient of 0.9976 where D_{12} is in cm^2/s and T is in Kelvin.

Yang, Hawkes, and Lindstrom have described (1) a method for the determination of diffusion coefficients of organic tracers in inorganic gases, using Knox and McLaren's "arrested-elution" variant (2) of the chromatographic method. Briefly, a narrow zone of tracer was injected into a stream of helium and carried to the center of a long, narrow tube. The flow was then stopped and the tracer was allowed to diffuse for a measured time. The flow was then restarted and the tracer was swept out of the column through a detector which recorded the concentration-time curve. This was approximately a Gaussian peak. The exact equation to the peak was derived and fitted to the data to give the value of D_{12} as the only fitted data point.

Supplying an initial input zone of negligible width required a specially designed valve. This failed repeatedly so that a new and more reliable inlet system seemed desirable.

Experimental Section

The apparatus and the methodology were similar to those of Yang and Hawkes (1) with the following modified injection system.

The diffusant (*n*-propane) was injected into a stream of helium by using a Loenco chromatographic injection valve. The mixture flowed through a Carle thermal conductivity detector whose output was observed on a strip-chart recorder. Near the top of the peak the flow was diverted into the main apparatus

Table I. Binary Molecular Diffusion Coefficient for $\text{C}_3\text{H}_8-\text{He}$

velocity of flow, cm/s	arrested time, s	temp, °C	D_{12}^* , cm^2/s	$10^3(\text{std error of } D_{12}^*)^a$
2.938	300	21.3	0.3005	1.0
3.060	300	30.3	0.3292	1.0
3.192	300	40.1	0.3601	1.0
3.461	300	51.0	0.4104	1.1
3.678	300	60.3	0.4365	1.5

^a From least-squares fit.

for a few carefully timed seconds. The input distribution of the tracer was therefore nearly rectangular and its width was precisely known. The input distribution was only 1% of the final distribution so that any errors in its measurement had negligible effect.

Calculations

The value of D_{12} was calculated from Lindstrom's equation (1) exactly as before. The result was corrected for the measured input distribution by assuming that the variance of the output distribution σ_{out}^2 was the sum of the variances of the input distribution and the distribution due to diffusion; i.e.

$$\sigma_{\text{out}}^2 = \sigma_{\text{in}}^2 + \sigma_{\text{diff}}^2 \quad (1)$$

Using the Einstein equation (3) $\sigma_{\text{diff}}^2 = 2Dt$ and the equation for a rectangular distribution $\sigma^2 = L^2/12$ where L is the length of the rectangular input in units of length gave

$$D_{12} = D_{\text{measured}} - L^2/(24t) \quad (2)$$

Comparison with Measurement by Yang and Hawkes

Two determinations of D_{12} at 18.8 °C for methane in helium gave an average value of $0.6431 \pm 0.0017 \text{ cm}^2/\text{s}$. Interpo-

Table II. Comparison of Experimental and Calculated Diffusion Coefficients for C₃H₈-He

temp, °C	D ₁₂ , cm ² /s		
	exptl	from eq 3	D ₁₂ (calcd) - D ₁₂ (exptl)
21.3	0.3005 ± 0.0010	0.3002	-0.0003
30.3	0.3292 ± 0.0010	0.3295	+0.0003
40.1	0.3601 ± 0.0010	0.3633	+0.0022
51.0	0.4104 ± 0.0011	0.4036	-0.0068
60.3	0.4365 ± 0.0015	0.4403	+0.0038

lation between Yang and Hawkes' (4) values of 15 and 20 °C gave $D_{12} = 0.6446 \text{ cm}^2/\text{s}$. The expected reliability of either determination (4) is 0.2% or 0.0013, so that a difference of 0.0015 between the two measurements is satisfactory.

Propane in Helium

The new measured data for propane in helium are shown in Table I. These were fitted to the equation $\log D_{12} = A + B$

log T by using a linear least-squares regression and gave

$$\log D_{12} = -8.1277 + 3.0805 \log T \quad (3)$$

with a correlation coefficient of 0.9976. The coefficients require D_{12} to be in cm²/s and T to be in Kelvin. The number of significant figures quoted is required because the fitting of the two numbers is highly correlated so that rounding of either requires adjustment of the other. Individually, they could not be relied upon to the implied precision.

The experimental and fitted values are shown in Table II.

Registry No. Propane, 74-98-6; helium, 7440-59-7.

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Sound Velocity in Formamide

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The speed of sound and the isentropic compressibility of formamide were measured by Brillouin spectroscopy in the temperature range from 293 to 373 K.

Introduction

The speed of sound and the isentropic compressibility of formamide were measured from 293 to 373 K by using Brillouin light scattering. The measurements were made as part of a study of sound velocity in associated liquids. Brillouin spectroscopy has been shown to provide accurate values of the speed of sound and isentropic compressibility in liquids (1). A survey of the literature revealed that no one has previously reported these values as a function of temperature or by Brillouin light scattering.

Experimental Section

Purification of Materials. Spectroscopic-grade formamide obtained from Aldrich was used without further purification. The formamide was repeatedly filtered through fritted glass with a pore size of 4.5–5.0 μm into a fluorescence cell. The filtration is necessary in order to remove dust from the formamide which may obscure the Brillouin peaks.

Measurement of the Speed of Sound. A description of the experimental setup has been previously published (2). An argon ion laser operated at 514.5 nm was focused onto the sample cell, which was thermostated to ±0.1 K. The scattered light was focused through a Fabry-Perot interferometer with a long focal length lens. Output of the interferometer was focused with a second lens through a pinhole onto a photomul-

Table I. Speed of Sound and Isentropic Compressibility of Formamide

T, K	ω _s , GHz	C _s , ms ⁻¹	κ _s , GPa
296.1	6.60	1661	0.320
300.3	6.53	1646	0.327
303.7	6.51	1641	0.329
306.9	6.45	1628	0.336
309.9	6.41	1619	0.340
316.8	6.38	1614	0.344
319.5	6.35	1607	0.348
326.9	6.27	1588	0.358
329.4	6.22	1577	0.364
332.1	6.18	1568	0.369
336.1	6.17	1567	0.370
339.1	6.14	1560	0.375
342.2	6.12	1557	0.377
345.1	6.10	1552	0.380
348.5	6.05	1541	0.387
353.1	6.00	1531	0.393
356.1	5.99	1528	0.396
359.3	5.96	1523	0.399
362.7	5.92	1514	0.405
365.9	5.90	1508	0.409
368.7	5.85	1497	0.416
372.0	5.82	1491	0.420

tiplier tube (PMT). The output of the PMT was digitized and stored on an LSI 11/03 minicomputer.

The free spectral range (FSR) was 19.34 GHz. The sound velocities are accurate to ±2% and the isentropic compressibilities to ±3%.

Results and Discussion

The speed of sound and isentropic compressibilities for formamide as a function of temperature are given in Table I. The experimentally measured quantity in Brillouin light scattering is