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Viscosity and Density of Methane + *cis*-Decalin from 323 to 423 K at Pressures to 140 MPa

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Dynamic viscosity (η) and density (ρ) data are reported for methane + *cis*-decahydronaphthaline (decalin) binary mixtures of 25, 50, and 75 mass% (74, 90, and 96 mol%) methane at three temperatures (323, 373, and 423 K) from saturation pressure to 140 MPa. A capillary tube viscometer was used for measuring the dynamic viscosity, with the density being calculated from measurements of sample mass and volume. The overall uncertainties in the reported data are 1.0 and 0.5% for the viscosity and density measurements, respectively.

KEY WORDS: binary mixtures; *cis*-decalin; decahydronaphthaline (decalin); density, high pressure; methane; viscosity.

1. INTRODUCTION

Accurate knowledge of the viscosity and density of fluids is crucial in numerous chemical and petroleum engineering processes. However, it is often not economically viable to make such measurements for a wide range of fluid compositions at different pressures and temperatures. Predictive models can provide a useful and more economical alternative to this problem, although the reliability of predictive techniques depends greatly on the accuracy and availability of experimental data.

The viscosity and density of hydrocarbon fluids play a major role in the flow characteristics of fluids in petroleum reservoirs and, hence, in the recovery of hydrocarbons. In field development planning, the number of wells and the location of each well are dependent on the viscosity of reservoir fluids. Unreliable viscosity predictions could result in operations

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either incurring unnecessary costs or failing to achieve production targets. Popular viscosity prediction models usually depend heavily on accurate density measurements [1]. Experimental density measurements are also very useful in calculating kinematic viscosity from absolute viscosity and density data; hence, many laboratories endeavor to include density measurements in their viscosity measurement program.

Recent advances in drilling and production technologies are facilitating exploration and production in deep formations that host hydrocarbon accumulations at high temperatures and pressures. However, there is a lack of experimental viscosity and density data available for the optimization and validation of numerical models when extending predictions to such extreme conditions. An ongoing 3-year research program, supported by the EU, has the aim of filling this gap. The main objective of the program is to provide reliable experimental density and viscosity data for binary, multicomponent, and real reservoir fluids under elevated pressure and temperature conditions. The generated data will be used by other partner institutions for the development and validation of predictive techniques.

In a previous paper [2], details of the experimental setup and test procedures for measuring the viscosity and density of hydrocarbon reservoir fluids at elevated temperatures and pressures were described. In addition, the test facilities were used to generate viscosity and density data for pentane and decane, which, when compared with literature data, demonstrated the reliability of the experimental equipment and test procedures. Finally, the density and viscosity of methane + methylcyclohexane mixtures were measured and reported. In this paper, experimental viscosity and density data on methane + *cis*-decalin at 25, 50, and 75 mass% (74, 90, and 96 mol%) methane are presented. The data cover a wide range of temperature and pressure conditions relevant to petroleum exploration and production (323 to 423 K, up to 140 MPa).

Methane + *cis*-decalin was chosen as a system containing a bicyclic saturated hydrocarbon. Decalin ($C_{10}H_{18}$) exists in petroleum reservoir fluids and is a widely used solvent in the production of paints, lacquers, waxes, and polishes. The density and viscosity of *trans*-decalin at atmospheric pressure have been reported in the literature [3]. No information is available on the viscosity and density of methane + *cis*-decalin systems for comparison with the data generated in this work.

2. APPARATUS AND TEST PROCEDURE

A detailed description of the experimental equipment and test procedures is presented elsewhere [2]. Viscosity measurements are conducted by means of a capillary tube viscometer, which is incorporated into the *PVT* facility. The experimental apparatus can be used to study the *PVT* phase behavior and properties of pure, multicomponent, synthetic, and real reservoir fluid systems with and without water. The current capabilities include measurements of dew point, bubble point, phase volumes, interfacial tension, density, and viscosity. The *PVT* facility can be used to conduct measurements from ambient conditions to a maximum pressure of 133 MPa at 524 K or 200 MPa at 474 K.

The capillary tube used in the viscosity measurements was 1480.3 cm long with an internal average diameter of 0.02994 cm. Two Quartzdyne pressure transducers (207 MPa; uncertainty, ± 0.02 MPa) monitor the system pressure as well as the differential pressure across the tube.

For viscosity measurements, the sample was passed through the capillary tube at three or four flow rates at each system pressure. At each flow rate, the differential pressure across the tube was measured. A number of repetitions at each flow rate were carried out. Generally, there was good agreement among different repetitions. The test was terminated when three consecutive measurements gave less than 1% uncertainty in the calculated viscosity. The test was repeated at a minimum of three or four flow rates. Again, there was excellent agreement in the calculated viscosities at different flow rates. The reported viscosity at each pressure is an average of the three or four readings. Measurements were made only at flow rates where laminar flow was established, i.e., for Reynolds numbers below 100.

The viscosity is calculated using the modified Poiseuille equation for the flow of compressible fluids through a tube. During viscosity measurements, the differential pressure across the capillary tube varied from about 0.007 to 0.2 MPa; hence, the fluid compressibility factor can be assumed to be constant along the tube, resulting in the following equations for isothermal compressible fluid flow:

$$q_1 = \frac{\pi D^4}{256 \eta L P_1} \times (P_1^2 - P_2^2) \tag{1}$$

$$\eta = \frac{\pi D^4}{256 q_1 L P_1} \times (P_1^2 - P_2^2) \tag{2}$$

where L is the length of the tube (in m), D is the tube diameter (in m), P_1 is the inlet pressure and P_2 is the outlet pressure (in Pa), η is the viscosity (in Pa.s), and q_1 is the fluid volumetric flow rate (in m^3) under the inlet pressure conditions. The impact of end-effect and kinetic-energy correction factors were found to be negligible, as a result of employing a long (1480.3-cm) capillary tube and using low flow rates, respectively. The effect of radial acceleration [4] was also calculated to be less than 0.2% by calculating the Dean number under maximum Reynolds number conditions.

The average radius of the tube was calculated from the measured volume and length of the tube. The effect of pressure and temperature on the capillary tube was calculated by neglecting the effect of temperature on the Young's modulus and on the Poisson's ratio. Considering the accuracy of measured parameters, the uncertainty in the reported viscosity was estimated to be about $\pm 1\%$ for viscosities in the range of those measured in this work. The error is due mainly to the contribution of the measured differential pressure.

The density was calculated from the mass and volume of the sample. The procedure was that the volume of the cell was measured accurately by filling it with mercury and weighing it on an accurate balance. The mass of the test fluid is measured directly by transferring it to the cell after an initial vacuum. Then mercury was pumped into the cell to achieve the desired pressure and the change in the sample volume was monitored. The density was calculated directly once the temperature and pressure had been stabilized. The full test procedure is described in detail elsewhere [2].

2.1. Test Fluids

Instrument-grade (99.995% pure) methane (CH_4; $MW_t = 16.04$ g/gmol) was purchased from Air Products, with 99% pure *cis*-decalin ($C_{10}H_{18}$; $MW_t = 138.25$ g/gmol) supplied by Aldrich.

3. RESULTS AND DISCUSSION

A series of viscosity and density measurements was made on three compositions (25, 50, and 75 mass% methane) of methane + *cis*-decalin at three temperatures (323, 373, and 423 K) and at pressures up to 140 MPa.

For each mixture, the various compositions were prepared gravimetrically, as reported in Tables I to III. The saturation pressure for each mixture at the three selected temperatures was measured using the visual capability of the experimental setup. The saturation pressure data for the three isotherms for the different fluid compositions are reported in Table IV.

The experimental viscosity data on methane + *cis*-decalin systems containing 25, 50, and 75mass% methane are presented in Tables I to III. The viscosity measurements were conducted at 323, 373, and 423 K for each system.

The results of the density measurements made on methane and *cis*-decalin at different compositions and temperatures are given in Tables V, VI, and VII. As with the viscosity data, the composition of the mixture is

Table I. Viscosity, η, of Methane (25.1 ± 0.4 mass%) + *cis*-Decalin

P (MPa)[a]	η (μPa·s)[b]	P (MPa)	η (μPa·s)	P (MPa)	η (μPa·s)
$T = (323 \pm 0.5)$ K		$T = (373 \pm 0.5)$ K		$T = (423 \pm 0.5)$ K	
58.75	288.2	50.47	180.4	48.69	129.7
64.32	298.6	55.51	191.6	56.09	140.2
72.53	317.7	70.71	217.4	72.25	161.0
85.77	349.0	86.95	243.6	88.51	184.3
102.79	387.7	105.54	273.7	103.26	202.8
119.81	428.5	120.16	297.7	121.99	228.4
138.24	479.7	138.00	332.5	137.69	247.3

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±1%.

Table II. Viscosity, η, of Methane (50.0 ± 0.2 mass%) + *cis*-Decalin

P (MPa)[a]	η (μPa·s)[b]	P (MPa)	η (μPa·s)	P (MPa)	η (μPa·s)
$T = (323 \pm 0.5)$ K		$T = (373 \pm 0.5)$ K		$T = (423 \pm 0.5)$ K	
69.19	93.1	59.50	64.2	51.53	47.4
86.60	102.7	71.78	71.6	69.12	57.6
96.47	110.2	86.09	81.0	86.07	67.5
104.00	115.6	103.29	90.5	103.21	75.7
117.33	124.2	120.30	100.3	120.82	84.2
138.71	135.6	137.70	111.7	138.12	93.1

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±1%.

Table III. Viscosity, η, of Methane (75.0 ± 0.1 mass%) + Decalin

P (MPa)[a]	η (μPa·s)[b]	P (MPa)	η (μPa·s)	P (MPa)	η (μPa·s)
$T = (323 \pm 0.5)$ K		$T = (373 \pm 0.5)$ K		$T = (423 \pm 0.5)$ K	
55.66	47.1	52.09	37.1	48.55	32.0
72.43	54.5	68.78	44.5	69.01	39.1
85.82	59.3	84.46	50.7	84.10	44.9
102.70	67.9	103.39	57.6	103.22	50.4
119.95	74.5	120.25	63.9	120.15	56.0
138.01	81.1	137.02	69.6	137.13	59.7

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±1%.

Table IV. Saturation Pressure (MPa) of Methane + *cis*-Decalin Systems

	Temperature (K)			
	323	373	423	
Methane concentration (mass%)[a]	Pressure (MPa)			Remark
25.1	54.95	47.85	43.28	Bubble point
50.0	66.19	55.57	47.02	Dew point
75.0	53.92	44.47	34.61	Dew point

[a] 25.1, 50.0, and 75.0 mass% methane is equivalent to a 74.3, 89.6, and 96.3 mol% methane concentration in the binary mixtures.

Table V. Density, ρ, of Methane (25.0 ± 0.4 mass%) + *cis*-Decalin

P (MPa)[a]	ρ (kg·m^{-3})[b]	P (MPa)	ρ (kg·m^{-3})	P (MPa)	ρ (kg·m^{-3})
$T = (323 \pm 0.5)$ K		$T = (373 \pm 0.5)$ K		$T = (423 \pm 0.5)$ K	
58.57	611	53.91	567	56.32	535
67.98	622	71.66	595	70.47	560
83.25	638	87.98	615	86.77	582
100.10	653	105.86	632	106.61	604
121.31	668	123.12	647	123.13	619
140.43	680	142.39	661	139.23	632

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±0.5%.

Table VI. Density, ρ, of Methane [(49.7 ± 0.2 mass%) at 323 K and (49.9 ± 0.2 mass%) at 373 and 423 K] + *cis*-Decalin

P (MPa)[a]	ρ (kg·m^{-3})[b]	P (MPa)	ρ (kg·m^{-3})	P (MPa)	ρ (kg·m^{-3})
$T = (323 \pm 0.5)$ K		$T = (373 \pm 0.5)$ K		$T = (423 \pm 0.5)$ K	
74.30	474	60.49	406	57.32	365
84.54	487	70.15	421	69.31	391
97.35	500	86.20	445	86.69	421
117.38	518	105.72	468	105.89	446
138.18	532	119.82	481	119.69	461
		140.12	498	139.07	479

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±0.5%.

Table VII. Density, ρ, of Methane (75.0 ± 0.1 mass%) + *cis*-Decalin

P (MPa)[a]	ρ (kg·m^{-3})[b]	P (MPa)	ρ (kg·m^{-3})	P (MPa)	ρ (kg·m^{-3})
T = (323 ± 0.5) K		T = (373 ± 0.5) K		T = (423 ± 0.5) K	
60.72	347	50.93	285	57.71	266
69.27	361	65.33	316	69.11	288
86.12	384	83.95	346	85.76	316
105.33	404	104.69	371	105.88	341
118.80	416	117.96	384	120.50	356
138.67	431	139.12	401	138.87	372

[a] Uncertainty, ±0.02 MPa.
[b] Uncertainty, ±0.5%.

presented with the density data. Figures 1 and 2 show the graphical presentation of Tables I and V, respectively.

4. CONCLUSIONS

Viscosity and density data for three methane + *cis*-decalin binary mixtures have been measured for a temperature range of 323 to 423 K, at pressures up to 140 MPa. The results should provide valuable information

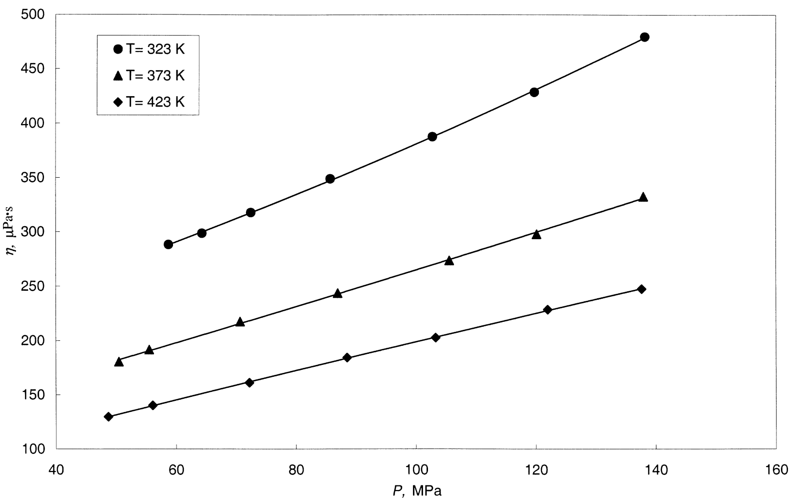

Fig. 1. Viscosity measurements for methane + *cis*-decalin (25.1 mass% methane) at different temperatures.

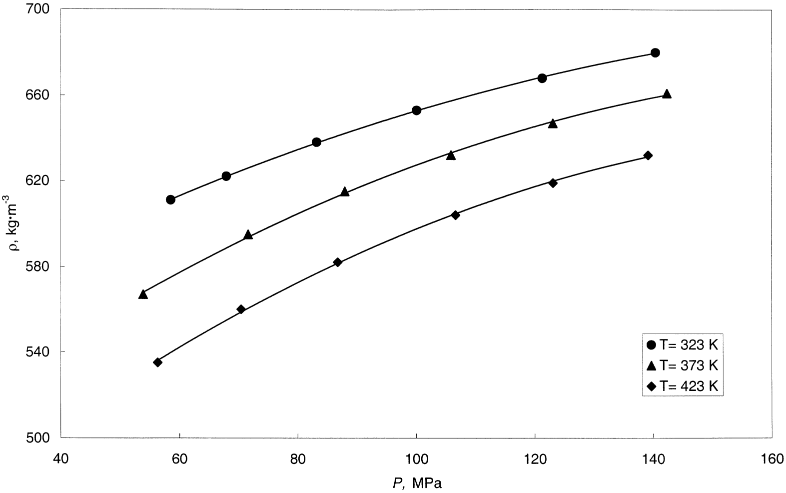

Fig. 2. Density measurements for methane + *cis*-decalin (25.0 mass% methane) at different temperatures.

for those assessing and developing viscosity and density prediction methods applicable to these extreme conditions.

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