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VISCOSITY OF HYDROCARBON LIQUIDS SATURATED WITH GAS

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ABSTRACT

Experimentally measured kinematic viscosities are reported for benzene, cyclohexane and n-heptane saturated with hydrogen, nitrogen and carbon dioxide. A Zeitfuchs cross-arm viscometer suspended in a unique design pressure cell was used to contain the viscometer at pressures from 200 to 1000 psia. Temperatures ranged from 75 to 200°F. Absolute viscosities were computed by use of an equation of state and a liquid density prediction method.

NOMENCLATURE

log = base 10 logarithm
t = temperature, °C

Greek Letters

ρ = density, gm/cc
μ = absolute viscosity, centipoise

Subcripts

t = temperature, °C
20 = at 20°C

INTRODUCTION

Viscosity is an important liquid characteristic. It influences equipment design and performance in many engineering areas. In many cases of industrial interest the liquid involved will be saturated with a gaseous component.

A survey of published literature reveals that most liquid viscosity data cover only limited ranges of temperature and pressure. Comparatively there is much more liquid viscosity data available for varying temperature than there is for varying pressure. Most of the available data is for pure liquids, or liquid mixtures. Few investigations have measured viscosities of liquids saturated with gaseous components.

This work reports the viscosity of n-heptane, cyclohexane and benzene saturated with carbon dioxide, hydrogen or nitrogen. Temperatures ranged from 70 to 200°F at pressures from 200 to 1000 psia. Experimental determinations were of kinematic viscosity. These were converted to absolute viscosities through compositions calculated by the Moshfeghian-Maddox computer program (8) for the Soave (12) version of the Redlich-Kwong (9) equation of state and Hankinson and Thomson (4) densities.

PREVIOUS WORK

Lewis (6) measured the viscosities of carbon tetrachloride, methyl alcohol, benzene acetone, and ether containing dissolved sulfur dioxide and chlorine. Most of the measurements were at room temperature and unspecified pressure. Lewis (6) concluded liquid viscosity varied non-linearly with the amount of gas dissolved in the liquid.

Simon and Graue (11) measured the viscosity of nine crude oils saturated with carbon dioxide. Temperatures ranged from 100 to 250°F at pressures to 2300 psia. At a given temperature the liquid viscosity decreased as the carbon dioxide pressure increased.

Bennett (3) measured the viscosity of n-nonane saturated with methane gas at temperatures from -30 to 78°F and pressures to 1200 psia with a wetted Zeitfuchs cross-arm viscometer. He showed that the wetted viscometer could be used for viscosity measurement. The viscosity of nonane decreased exponentially with increasing pressure (increasing methane content) at low temperatures, but approached a linear variation at higher temperatures. Bagzis (2) Used n-decane, n-hexane and a mixture of n-butane and n-decane saturated with methane. Rudolf (10) used lean absorber oils saturated with methane at temperatures from -26 to 77°F. The observations of Bagzis (2) and Rudolf (10) were in agreement with Bennett(3).

EXPERIMENTAL PROCEDURE

Figure 1 shows a schematic diagram of the experimental apparatus used in this investigation. The pressure distribution system served to introduce the test gas into the pressure cell, control the liquid level in the viscometer and measure total pressure in the cell. The in-line pressure controller and the Ruska pump were used to control the amount of liquid in the viscometer. Vacuum prior to introduction of the sample was produced by a Duo-Seal pump and measured by an Ascroft-American Duragauge graduated in 0.5 inch of mercury increments from 0 to 30 inches of mercury vacuum.

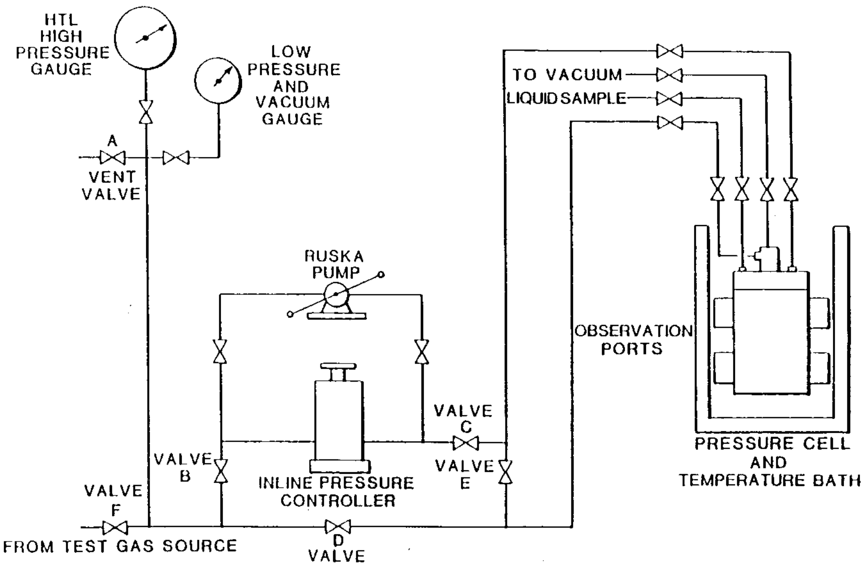

Fig. 1: Schematic diagram of experimental apparatus

The heart of the experimental equipment was a Zeitfuchs cross-arm capillary viscometer mounted in the high pressure (3000 psia maximum) cell with view ports that permitted viewing the liquid level and also the etched lines demarcating the fixed volume of fluid to flow. Observations inside the cell were made through quartz windows by use of a 14 power cathetometer. Cleanliness of the viscometer was paramount and it was triple washed before each use with absolute ethanol, triple distilled water and acetone and then dried by blowing with dehumidified, filtered air.

A Zeitfuchs Cross-Arm Capillary Viscometer is shown schematically in Figure 2. In conventional use the reservoir of the viscometer is filled to the scribed line with liquid. A slight pressure is applied to the surface of the liquid to initiate flow through the capillary and the time required to fill the measuring bulb is determined. From a viscometer constant determined by using a liquid of known viscosity, the kinematic viscosity of the test liquid is computed. The viscometer is then removed from service, thoroughly cleaned and dried to make it ready for another viscosity determination.

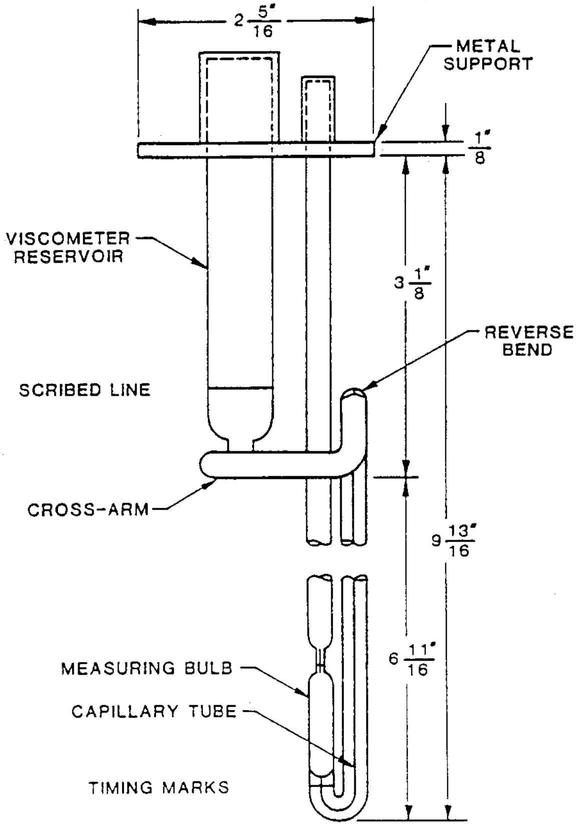

Fig. 2: Schematic diagram displaying dimensions of Zeitfuchs cross-arm viscometer

The viscosity determinations reported here required two modifications in the conventional procedure for viscosity measurement using the Zeitfuchs Cross-Arm Viscometer. The viscometer was suspended in a pressure cell to allow equilibration under high pressures of solute gas. By carefully using small pressure differences to return the liquid from the measuring bulb to the reservoir, multiple measurements were made at a given temperature and pressure without cleaning the viscometer. Bennett (3) proved conclusively that measurements made using a "wetted" Zeitfuchs viscometer in this manner were reproducible and accurate. Bagzis (2), Rudolf (10) and McCoy (7) confirmed and extended this portion of Bennett's work.

The viscometer was calibrated using distilled water. Absolute viscosities for water were computed using the equation of Kestin, et al. (5):

$$\log\left(\frac{\mu_t}{\mu_{20}}\right) = 1.2378 - \left[1.303\left(10^{-3}\right)(20-t)\right] + \left[3.06\left(10^{-6}\right)(20-t)^2\right] + \left[2.55\left(10^{-8}\right)(20-t)^3\right] \qquad (1)$$

Equation (1) applies over the temperature range -8 to 150 °C at atmospheric pressure. Conversion of absolute to kinematic viscosity for water was made using the water density equation of Abul-Hamayal (1):

$$\rho = 0.999986 + \left[0.1890\left(10^{-4}\right)(t)\right] - \left[0.5886\left(10^{-5}\right)\left(t^2\right)\right] + \left[0.1548\left(10^{-7}\right)\left(t^3\right)\right] \qquad (2)$$

Using properties from equations (1) and (2), the calibration constant for the Zeitfuchs Viscometer was determined to be 1.1830 X 10^{-3} as shown in Table 1. With this calibration constant the viscosity of n-heptane was determined at three different temperatures and atmospheric pressure. The comparison with API Project 44 (13) values of viscosity for n-heptane in Table 2 shows the measured values are within 1% of the API accepted values.

Table 1: Zeitfuchs Viscometer Calibration Using Distilled Water

Temperature °F	Time sec.	Absolute Viscosity cs	Density gm/cc	Kinematic Viscosity cs	Calibration Constant 1000x(cs/sec)
79.82	725.11	0.8594	0.9966	0.8623	1.1892
88.10	665.50	0.7778	0.9953	0.7815	1.1743
98.10	588.78	0.6951	0.9935	0.6696	1.1882
119.60	479.45	0.5595	0.9887	0.5659	1.1803
				Avg. :	1.1830

Table 2: Atmospheric Viscosity of Heptane Using Wetted Zeitfuchs Viscometer

Temperature °F	Experimental Kinematic Viscosity cs	API 44 Kinematic Viscosity cs	Error Percent
77.41	0.5859	0.5811	-0.80
98.46	0.5204	0.5255	0.90
124.08	0.4658	0.4696	0.38

Experimental determinations of viscosity were made for hydrocarbon liquids saturated with gases. The efficacy of the wetted wall viscometer has been shown, but the fact that the liquid will saturate with gas has not. Evidence for this is shown in Table 3. After several hours in the pressure cell, bulb fill time for the liquid approaches a highly reproducible value. Note that the first fill for any time is not representative. Apparently there is some drying of the viscometer capillary walls during times of inactivity.

Table 3: Proof of Steady State for N-Heptane in Carbon Dioxide

Time	Run No.	Temperature millivolts	Temperature °F	Pressure psig	Flow Time min:sec
1330	1	4.840	217.758	310	3:05.67
	2	4.840	217.758	310	3:28.00
	3	4.840	217.758	310	3:22.24
	4	4.841	217.802	310	3:45.55
	5	4.841	217.802	310	3:12.50
1530	1	4.840	217.758	310	3:45.37
	2	4.840	217.758	310	4:18.12
	3	4.840	217.758	310	4:06.42
	4	4.840	217.758	310	4:07.17
	5	4.841	217.802	310	4:11.35
1820	1	4.840	217.758	310	4:11.99
	2	4.840	217.758	310	4:07.13
	3	4.840	217.758	310	4:09.30
	4	4.841	217.802	310	4:10.30
	5	4.842	217.844	310	4:10.00
	6	4.840	217.758	310	4:10.26

RESULTS AND DISCUSSION

Experimental viscosity measurements were made for benzene, cyclohexane and n-heptane saturated with hydrogen, nitrogen and carbon dioxide. Tables 4, 5 and 6 show viscosities saturated with hydrogen, Tables 7, 8 and 9 with

Table 4: Viscosity of Benzene Under Hydrogen Pressure

H_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction H_2
215	77.57	0.7099	0.8669	0.6154	0.0071
	99.77	0.6101	0.8542	0.5211	0.0077
	135.48	0.4864	0.8332	0.4053	0.0085
	163.14	0.4385	0.8165	0.3580	0.0091
415	77.57	0.7271	0.8643	0.6284	0.0137
	99.77	0.6113	0.8512	0.5204	0.0148
	135.48	0.5043	0.8297	0.4184	0.0165
	163.14	0.4389	0.8125	0.3566	0.0179
615	77.57	0.7289	0.8616	0.6281	0.0201
	99.77	0.6225	0.8483	0.5281	0.0217
	135.48	0.5036	0.8262	0.4161	0.0244
	163.14	0.4411	0.8085	0.3566	0.0265

Table 5: Viscosity of Cyclohexane Under Hydrogen Pressure

H_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction H_2
215	99.13	0.9650	0.7590	0.7324	0.0108
	129.20	0.7950	0.7428	0.5905	0.0117
	148.07	0.7135	0.7325	0.5226	0.0122
	186.40	0.5625	0.7108	0.3998	0.0130
415	99.13	0.9723	0.7559	0.7349	0.0209
	129.20	0.8142	0.7393	0.6020	0.0226
	148.07	0.7300	0.7286	0.5319	0.0237
	186.40	0.5684	0.7061	0.4013	0.0259
615	99.13	0.9750	0.7528	0.7340	0.0306
	129.20	0.8265	0.7358	0.6081	0.0333
	148.07	0.7398	0.7246	0.5361	0.0350
	186.40	0.5720	0.7014	0.4012	0.0384

Table 6: Viscosity of Heptane Under Nitrogen Pressure

N_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction N_2
215	97.02	0.5300	0.6680	0.3540	0.0238
415		0.5572	0.6672	0.3718	0.0453
615		0.5655	0.6663	0.3768	0.0659
815		0.5785	0.6652	0.3848	0.0857
1015		0.5821	0.6641	0.3866	0.1049
215	166.90	0.3786	0.6319	0.2392	0.0245
415		0.4093	0.6302	0.2579	0.0472
615		0.4164	0.6282	0.2616	0.0691
815		0.4200	0.6261	0.2630	0.0903
1015		0.4224	0.6239	0.2635	0.1108

nitrogen and Tables 10, 11 and 12 with carbon dioxide. Tables 4 through 12 show kinematic and absolute viscosities and the calculated gas component concentration in the liquid phase.

Table 7: Viscosity of Benzene Under Nitrogen Pressure

N_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction N_2
215	77.82	0.6902	0.8683	0.5993	0.0095
	125.64	0.5128	0.8409	0.4312	0.0100
	149.79	0.4442	0.8267	0.3672	0.0102
	184.94	0.3783	0.8053	0.3046	0.0104
415	77.82	0.7059	0.8671	0.6121	0.0181
	125.64	0.5278	0.8393	0.4430	0.0192
	149.79	0.4594	0.8248	0.3789	0.0198
	184.94	0.3862	0.8031	0.3101	0.0207
615	77.82	0.7164	0.8658	0.6203	0.0265
	125.64	0.5339	0.8377	0.4473	0.0282
	149.79	0.4671	0.8230	0.3844	0.0292
	184.94	0.3949	0.8008	0.3163	0.0307

Table 8: Viscosity of Cyclohexane Under Nitrogen Pressure

N_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction N_2
215	96.17	0.9852	0.7626	0.7514	0.0143
	125.06	0.7824	0.7475	0.5848	0.0145
	166.48	0.6253	0.7247	0.4531	0.0147
	214.90	0.4715	0.6955	0.3279	0.0157
415	96.17	0.9957	0.7616	0.7583	0.0273
	125.06	0.7980	0.7460	0.5953	0.0279
	166.48	0.6188	0.7229	0.4473	0.0288
	214.90	0.4755	0.6944	0.3302	0.0299
615	96.17	0.0073	0.7605	0.7660	0.0399
	125.06	0.8210	0.7447	0.6114	0.0409
	166.48	0.6288	0.7211	0.4534	0.0425
	214.90	0.4762	0.6918	0.3294	0.0447
815	96.17	0.0040	0.7593	0.7623	0.0522
	125.06	0.8210	0.7432	0.6102	0.0535
	166.48	0.6332	0.7192	0.4554	0.0559
	214.90	0.4784	0.6891	0.3297	0.0592

Table 9: Viscosity of Heptane Under Hydrogen Pressure

H_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction H_2
215	73.77	0.6053	0.6767	0.4096	0.0165
	114.84	0.4936	0.6560	0.3238	0.0182
	148.20	0.4588	0.6386	0.2930	0.0195
	186.23	0.3795	0.6180	0.2345	0.0210
415	73.77	0.6128	0.6736	0.4128	0.0315
	114.84	0.5130	0.6523	0.3346	0.0348
	148.20	0.4632	0.6342	0.2938	0.0376
	186.23	0.3901	0.6126	0.2390	0.0408
615	73.77	0.6200	0.6704	0.4157	0.0460
	114.84	0.5253	0.6484	0.3406	0.0590
	148.20	0.4783	0.6297	0.3012	0.0509
	186.23	0.4100	0.6071	0.2489	0.0600

Table 10: Viscosity of Benzene Under Carbon Dioxide Pressure

CO_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction CO_2
215	68.29	0.5647	0.8958	0.5059	0.1302
	105.60	0.4637	0.8681	0.4025	0.0989
	140.81	0.4183	0.8436	0.3529	0.0798
	184.51	0.3554	0.8141	0.2893	0.0634
415	68.29	0.4306	0.9169	0.3948	0.2641
	105.60	0.4228	0.8811	0.3725	0.1947
	140.81	0.3926	0.8526	0.3347	0.1567
	184.51	0.3412	0.8196	0.2796	0.1263
615	68.29	0.2817	0.9411	0.2651	0.4345
	105.60	0.3563	0.8939	0.3185	0.2955
	140.81	0.3417	0.8608	0.2941	0.2343
	184.51	0.3156	0.8245	0.2602	0.1889

Table 11: Viscosity of Cyclohexane Under Carbon Dioxide Pressure

CO_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction CO_2
225	88.13	0.8183	0.7931	0.6490	0.1393
	116.10	0.7075	0.7726	0.5466	0.1156
	157.91	0.5700	0.7442	0.4242	0.0913
	174.20	0.5143	0.7335	0.3772	0.0839
425	88.13	0.6423	0.8169	0.5247	0.2686
	116.10	0.6143	0.7901	0.4853	0.2205
	157.91	0.5227	0.7553	0.3948	0.1706
	174.20	0.4858	0.7436	0.3612	0.1617
615	88.13	0.5200	0.8413	0.4375	0.4035
	116.10	0.5089	0.8065	0.4105	0.3224
	157.91	0.4962	0.7663	0.3803	0.2532
	174.20	0.4284	0.7524	0.3223	0.2345

Table 12: Viscosity of Heptane Under Carbon Dioxide Pressure

CO_2 Pressure psia	Temp. °F	Kinematic Viscosity cs	Density g/cc	Absolute Viscosity cp	Mole Fraction CO_2
325	71.87	0.4640	0.7334	0.3403	0.2730
	126.91	0.3975	0.6852	0.2724	0.1945
	169.65	0.3473	0.6540	0.2272	0.1602
	215.30	0.2960	0.6225	0.1842	0.1355
525	71.87	0.3801	0.7713	0.2932	0.4488
	126.91	0.3535	0.7043	0.2490	0.3089
	169.65	0.3060	0.6668	0.2040	0.2548
	215.30	0.2745	0.6307	0.1731	0.2180
715	71.87	0.3420	0.8213	0.2809	0.6735
	126.91	0.3154	0.7219	0.2277	0.4158
	169.65	0.2850	0.6778	0.1932	0.3413
	215.30	0.2590	0.6372	0.1650	0.2932

Calculating absolute viscosity requires liquid densities, which require liquid phase composition. The liquid phase composition was calculated using the Moshfeghian and Maddox (8) version of the Soave modification (12) of the Redlich - Kwong equation of state (9). The density of the gas saturated liquid was calculated from the composition using the Hankinson - Thomson (4) relations. Because of low solubility in all three hydrocarbons, hydrogen and nitrogen exert minimal influence on liquid phase viscosity. In all cases for those two gases the temperature change creates much the largest change in liquid viscosity. In the case of both hydrogen and nitrogen slight reverse solubility behavior is observed. Carbon dioxide is an order of magnitude more soluble in the liquid hydrocarbons and the dissolved gas causes a significant lowering of the viscosity.

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