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THE GAS INDUSTRY'S OWN EDUCATIONAL AND RESEARCH FACILITY

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Improved High Pressure Capillary Tube Viscometer

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ABSTRACT

An apparatus and a procedure for determining the viscosity behavior of hydrocarbons at pressures up to 10,000 psia and temperatures between 77 and 400° F are described. The equipment is suitable for measuring viscosity of either the liquid or vapor phases or the fluid above the two-phase envelope for systems exhibiting retrograde phenomena, according to the phase state of the system within these ranges of temperature and pressure. Equations are developed for calculation of viscosity from the experimental measurements, and new data for the viscosities of ethane and propane at 77° F are reported.

INTRODUCTION

With the advent of higher pressures and temperatures in industrial processes and deep petroleum and natural gas reservoirs, demand has increased for accurate values of physical properties of hydrocarbons under these conditions. Proportionately, more frequent occurrence of natural gas and condensate-type fluids is encountered as fluid hydrocarbons are discovered at greater depths. This increases the importance, to the reservoir engineer, of being able to predict accurately the physical properties of light hydrocarbon systems in the dense-gas and light-liquid phase states.

Reliable gas viscosity data are limited primarily to measurements made on pure components near ambient temperature and at low pressures. Few investigations have been reported for high pressures, and except for methane, data on light hydrocarbons are subject to question. This is demonstrated by the large discrepancy between sets of data on the same component reported by different investigators. For mixtures in the dense gas and light liquid regions and for fluids exhibiting retrograde behavior there are very few published experimental data.

Viscosity data for methane have been reported by Bicher and Katz,¹ Sage and Lacey,¹² Comings, *et al.*,⁵ Golubev,⁸ and Carr,² with good agreement among the last three sets of data. Comings, Golubev and Carr utilized capillary tube instruments for which the theory of fluid flow is well established. The theory permits calculation of the viscosity directly from the experi-

mental data and dimensions of the instrument alone. Sage and Lacey, and Bicher and Katz used rolling-ball viscometers. The theory of the rolling-ball viscometer has not been completely established, and these instruments presently require calibration by use of fluids of known viscosity behavior before viscosities of test fluids can be measured. To obtain accurate data it is necessary that the rolling-ball viscometers be calibrated by use of fluids of density and viscosity similar to the test fluids, a difficult selection for the gas phase.

From the methane data and experimental tests on various natural gases, Carr developed a correlation for predicting the PVT behavior of light natural gases.^{2,3,4} This correlation was based on data for a very limited composition range; its application to rich gases and condensate fluids is questionable.

The object of this investigation is to develop an instrument which can be used to obtain viscosity data at reservoir temperatures and pressures, for rich gases, condensate-type systems above the two-phase envelope and light liquid mixtures. These data will be used in an effort to develop correlations to represent the viscosity behavior of these fluids.

APPARATUS

In a previous viscosity study Carr² utilized a modified Rankine capillary viscometer configuration,¹¹ Fig. 1. In this instrument the gas to be tested is forced through the capillary tube in laminar flow by motion of a mercury pellet in the fall tube, the measured displacement time being that required for the mercury slug to move between the brass timer rings. The viscometer is constructed of glass and mounted in a steel pressure vessel. The test gas pressure in the viscometer is balanced by an inert gas (usually nitrogen) in the vessel.

Excellent results have been obtained with instruments of this type, with Carr² and Comings⁵ reporting reproducibilities of 99.5 to 99.3 per cent and an estimated absolute accuracy of 99 per cent. However, these instruments have limitations which have precluded their use for liquids. The need for maintaining a balance between pressures of the test fluid and inert gas in the viscometer vessel presents operating problems, and requires charging the test fluid to the viscometer very slowly. The principle drawback to the Rankine unit is behavior of the mercury slug which provides the pressure differential across the capillary. When even trace quantities of propane or heavier hydrocarbons are present in the test gas, the mercury tends to subdivide

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¹References given at end of paper.

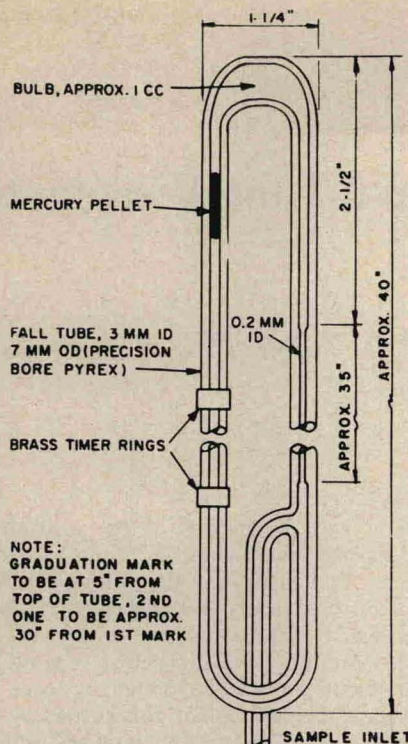


FIG. 1—DETAILS OF RANKINE-TYPE CAPILLARY TUBE VISCOMETER.

into small pellets which cannot be made to recombine in the fall tube; this necessitates disassembling and cleaning the viscometer.

A study was made of the major types of viscometers reported in the literature,⁶ and it was decided to base the new design on the principle of the Rankine viscometer, i.e., transpiration of the fluid through a capillary tube, but to devise a new method for providing the pressure differential. Accordingly, glass models of several different configurations were constructed and tested until a relatively simple apparatus was developed.

A schematic diagram of the new viscometer and associated equipment is presented in Fig. 2. The principle components of the instrument are: mercury receiver, K; mercury reservoir, N; high pressure swivel joints, H; glass capillary tube, L; capillary tube jacket, M; by-pass valve, J; and the mercury flow tube. The mercury receiver is fixed in position while the reservoir is free to move in a vertical plane.

The mercury reservoir and receiver were machined from 3.125-in. diameter 316 stainless steel round stock, and with tops in place are 3.688-in. high. Both vessels were bored to give chambers 1.0000 ± 0.0002-in. diameters by 1.625-in. deep, and are equipped with O-ring closures. The receiver body is tapped near the bottom

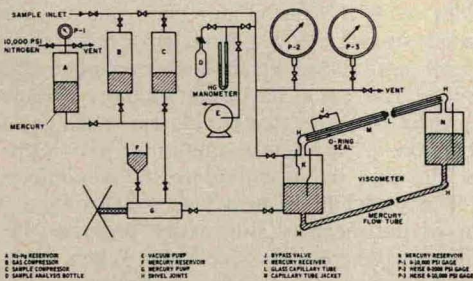


FIG. 2—SCHEMATIC DIAGRAM OF NEW HIGH PRESSURE CAPILLARY TUBE VISCOMETER.

for two 0.25-in. high pressure "Amico"-type fittings, and two fittings are also provided in the caps. The receiver cap is also drilled to receive two electrodes. The reservoir has only one fitting in the body and one in the cap, and the cap is drilled for only one electrode.

The simplified electrode assembly,¹⁵ shown in Fig. 3, was developed to facilitate accurate location of the electrodes, and minimize chance of dimension change during operation of the apparatus. This is very important since the calculated volume of fluid displaced and the change in driving force during the run are both based on the measured distance between receiver electrode tips. The long receiver electrode extends to within about 0.5 in. of the bottom, thus providing a volume below the electrode to permit flow conditions to stabilize before flow timing begins.

When the rising mercury surface contacts the long receiver electrode the timer starts, and when the mercury contacts the short electrode, the timer stops. The spacing between the electrode tips, measured to ± 0.0003 cm by use of a microcomparator, is about 1.45-cm (9/16 in.). The volume above the short electrode was provided to contain the mercury in the flow tube and eliminate the possibility of mercury getting into the upper swivel joint or the capillary tube. The reservoir electrode extends to within about 0.125 in. of the bottom, and is used in adjusting the volume of mercury in the system.

The receiver is rigidly fixed in place with the reservoir free to move in an arc of radius equal to the length of the mercury flow tube. The flow tube and capillary jacket are attached to the receiver and reservoir by 0.25-in. high pressure swivel joints, illustrated schematically in Fig. 4. The reservoir will remain upright as it is raised, since the mercury flow tube and the capillary tube jacket assembly act as parallel linkages. In this manner the pressure differential across the capillary tube (which is equivalent to the difference in levels of the mercury surfaces in the reservoir and the receiver) can be varied from 3 to 40 cm by raising the mercury reservoir to predetermined positions.

The mercury flow tube is a length of 0.25-in. OD, 0.125-in. ID, stainless steel tubing, rated for 15,000-psi working pressure. The capillary tube jacket is 9/16-in. OD, 0.25-in. ID, stainless steel tubing, also rated for 15,000 psi. The annulus between the capillary tube and the tube jacket is utilized as a fluid by-pass to permit rapid transfer of the test fluid between the reservoir and the receiver. This provided a means for rapid return of the mercury to the reservoir after a test run, and also can be used to insure homogeneity of the test fluid. The annulus is sealed at the receiver end by a 7/32-in. ID, 11/32-in. OD, O-ring fitted into a standard 9/16-in. tee utilizing the tube jacket as a follower. The by-pass valve is connected across the O-ring annulus seal, as shown in detail in Fig. 5.

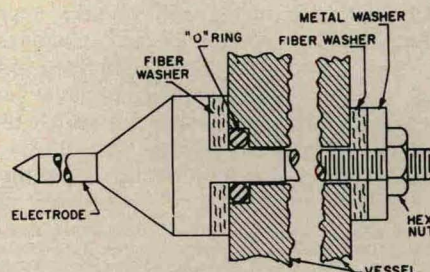


FIG. 3—ELECTRODE ASSEMBLY, O-RING TYPE.

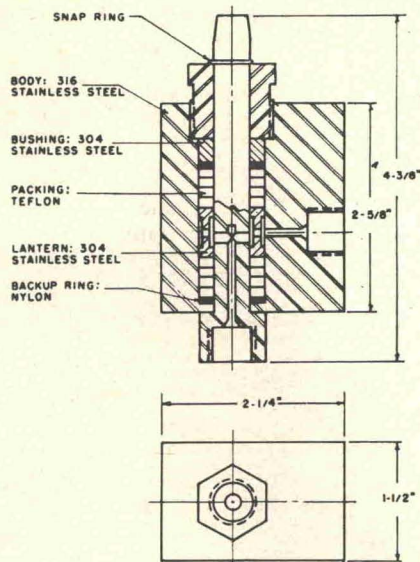


FIG. 4—AUTOCRAVE 0.25-IN. HIGH PRESSURE SWIVEL JOINTS.

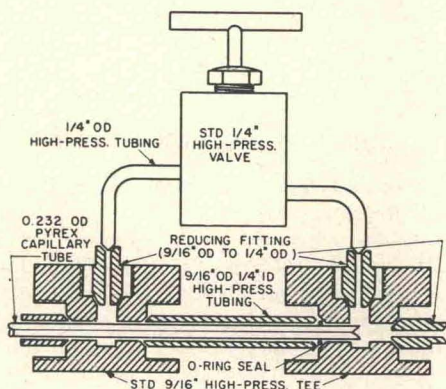


FIG. 5—DETAILS OF CAPILLARY TUBE SEAL AND BY-PASS VALVE ASSEMBLY.

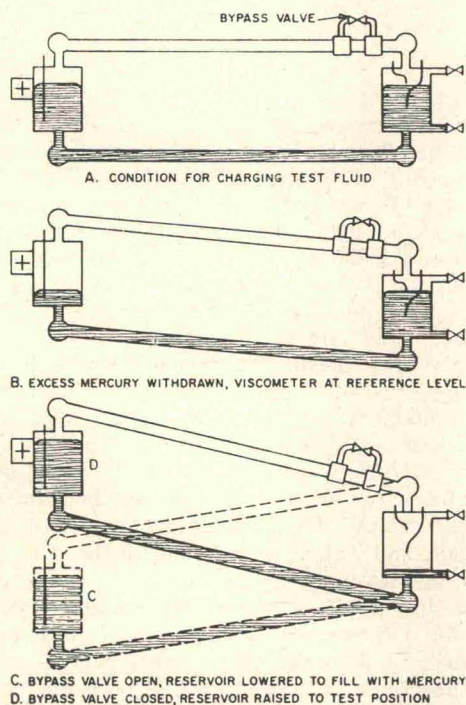


FIG. 6—RELATIVE RESERVOIR AND RECEIVER POSITIONS DURING CHARGING AND TESTS.

TABLE 1—SUMMARY OF CAPILLARY TUBE CALIBRATION DATA

	Tube No. 13	Tube No. 14
Length, L (cm)	90.070	91.165
r_1 by Fisher ⁷ method, cm	0.010876	0.012088
r_1 by gravimetric method, cm	0.010877	0.012092
$\int_0^L \frac{dL}{r_1^4}$ by Fisher ⁷ method, cm^{-3}	64.380×10^8	42.695×10^8
$\int_0^L \frac{dL}{r_1^4}$ by gravimetric method, cm^{-3}	64.359×10^8	42.648×10^8

The Pyrex capillary tubes were obtained as a special draw. The tubes are from 0.021- to 0.025-cm bore by 0.5- to 0.6-cm OD, and from 90 to 95 cm in length. The ends of the capillary tubes are flared slightly to provide a streamlined entrance and exit with consequent reduction in end-effects. Average internal radii of the capillary tubes were determined gravimetrically, and the variation of radius along the tubes was determined by a graphical integration technique proposed by Fisher⁷ and described in detail by Carr.² A summary of calibration data for two of the capillary tubes which have been used is given in Table 1.

EXPERIMENTAL PROCEDURE

Prior to assembly of the viscometer all parts were thoroughly cleaned and dried. The capillary tube was cleaned with chromic acid solution followed by flushing with water, acetone and ether. The metal parts were degreased and rinsed in acetone.

The receiver electrode spacing was measured and the viscometer assembled, except for the top of the mercury reservoir. Clean, dry mercury was added to the system until approximately 1/3-in. of mercury remained in the reservoir when the mercury surface in the receiver just made contact with the short electrode, the top was then bolted in place. The technique of charging the mercury to the viscometer ensured that sufficient mercury was present in the system for proper operation.

After the assembled viscometer had been evacuated, the system was pressured to about 1,000 psi for a leak test. A 5,000-psi deadweight gauge measured pressure to the nearest pound. Oil in the deadweight gauge was separated from the test fluid in the viscometer by a diaphragm-type pressure-balance indicator. The viscometer system was considered pressure tight when no pressure change could be detected with the diaphragm indicator in one hour.

After any leaks were sealed the nitrogen was vented and the unit evacuated and charged with test fluid. The remaining operations are best described by reference to Fig. 6. Prior to charging the viscometer with test fluid the reservoir level was varied until the mercury in the receiver made contact with the short electrode as in A, Fig. 6. This provided a means for monitoring the position of the mercury in the unit during the charging operation. The by-pass valve was open during evacuation and charging.

The volume of mercury in the viscometer was adjusted by slowly draining mercury from the pressurized system. As mercury was removed the reservoir was raised to keep the mercury in contact with the short electrode. Withdrawal was stopped when the reservoir electrode tip was still submerged 2 to 4 mm and the short electrode just made contact with the mercury in the receiver, as in B, Fig. 6. The by-pass valve was open during mercury withdrawal and determination of the reference level.

The cathetometer reading for the scribed crosshair of the reservoir target is taken at this point and represents the reference level for calculating mercury driving forces used in the tests.

With the quantity of mercury in the system properly adjusted and the reservoir target reference level measured, the instrument was ready for viscosity measurements. The reservoir was lowered to permit most of the mercury in the system to accumulate there, as in C, Fig. 6, and the by-pass valve closed. The reservoir was raised to run position, as in D, Fig. 6. The cathetometer reading was set to provide a pre-selected pressure differential across the capillary tube, and the reservoir height carefully adjusted each time until the target was aligned with the cathetometer cross hairs. In this manner multiple runs could be made at the same driving force, which resulted in the variation in flow times being a direct comparison of data reproducibility.

The electrical circuit utilized to control the timer and indicate mercury levels is shown in Fig. 7. Switch S_5 , a five-pole rotary gang switch, has three positions: off, test and run. With S_5 in test position and toggle switches S_1 and S_2 closed, green lights on the controller indicate when mercury is in contact with the reservoir electrode or the short receiver electrode. With S_5 in the run position and S_2 and S_3 closed, the controller will close R_3 when mercury contacts the long receiver electrode, starting the timer, and will close R_2 when mercury contacts the short electrode, stopping the timer. Relay R_3 can be by-passed to insure dependable timer operation by closing S_4 after the timer has started, then R_3 can be released by opening S_4 .

Typical experimental data for nitrogen are given in Table 2. The reproducibility of the flow time proved an excellent indication of condition of the viscometer. Whenever flow time variations consistently exceeded 0.5 per cent, it was time to disassemble and clean the apparatus. Occasionally erratic timer readings were obtained (as illustrated in Table 2) far removed from the average of three or more repeat runs; these points were discarded before averaging. Generally, variation of the measurements was ± 0.2 per cent or less from the average value.

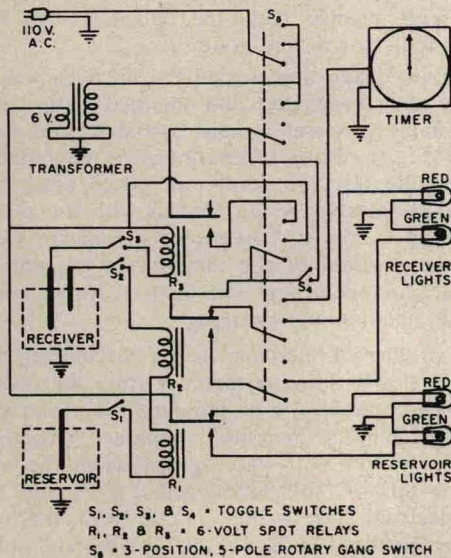


FIG. 7—WIRING DIAGRAM FOR TIMER CONTROL AND REFERENCE LEVEL CHECK.

TABLE 2—TYPICAL EXPERIMENTAL DATA ON NITROGEN

June 2, 1957, data set 6			June 3, 1957, data set 7		
Capillary 14			Capillary 14		
Temp. = 21.7°C			Temp. = 23.4°C		
Press. = 1,000.5 psig			Press. = 1,000.5 psig		
Bar. = 29.66 Hg			Bar. = 29.45 Hg		
Elec. spac. = 1.375 cm			Elec. spac. = 1.375 cm		
Run No.	Cathetometer Reading (cm)	Time (seconds)	Run No.	Cathetometer Reading (cm)	Time (seconds)
Base Line	67.770	—	Base Line	67.525	—
1	75.150	127.6	1	74.905	128.5
2	75.150	127.5	2	74.905	128.5
3	75.150	127.6	3	74.905	128.5
Avg	75.150	127.6±0.0	Avg	74.905	128.5±0.0
4	74.215	143.3	*4	73.975	145.8*
5	74.215	143.1	5	73.975	143.8
6	74.215	143.2	6	73.975	144.1
Avg	74.215	143.2±0.1	7	73.975	144.2
7	73.120	166.7	8	73.975	144.1
8	73.120	167.1	Avg	73.975	144.1±0.1
9	73.120	166.9	9	72.880	167.9
Avg	73.100	166.9±0.1	10	72.880	168.0
10	72.070	198.5	11	72.880	168.0
11	72.070	198.5	Avg	72.880	168.0±0.0
12	72.070	198.7	12	71.825	200.3
Avg	72.070	198.6±0.1	*13	71.825	171.3*
13	71.215	235.3	14	71.825	200.1
14	71.215	235.3	15	71.825	200.0
15	71.215	235.5	Avg	71.825	200.1±0.1
Avg	71.215	235.4±0.1	16	70.970	237.4
16	70.335	291.3	17	70.970	236.3
17	70.335	298.8	18	70.970	237.0
18	70.335	291.6	19	70.970	235.5
19	70.335	292.5	Avg	70.970	236.6±0.7
20	70.335	292.4	*20	70.090	324.3*
Avg	70.335	291.9±0.4	21	70.090	295.2
Base check	67.770	—	22	70.090	295.2
			23	70.090	294.8
			Avg	70.090	295.1±0.2
			Base check	67.540	—

*Omitted from average

RESULTS AND DISCUSSION

The theory of fluid flow through capillary tubes is very well developed. Equations have been derived for calculating the viscosity from experimental measurements, based on isothermal, laminar fluid flow in the capillary tube. The correlations for end effects, establishment of parabolic velocity distribution, compressibility of the test fluid and pressure losses in the mercury flow system are described in detail in the Appendix. The final equation used to calculate the results presented is as follows.

$$\mu_r = \frac{\pi g r_c^4 \theta}{8 L_c V_D} \left[\frac{\Delta h_{1m} (\rho_m - \rho_r) g}{g_c} \frac{\beta \rho_r V_D^2}{g \pi r_c^4 \theta^2} \right] \frac{L_m r_c^4 \mu_m}{r_m^4 L_c} \quad (1)$$

Initial tests of the viscometer were made by use of nitrogen at pressures from 200 to 1,000 psig at room temperature. Results are presented in Fig. 8, with the viscosity data of Michels and Gibson¹⁰ included for comparison. To determine data reproducibility, a series of eight sets of data (31 separate tests) were made at a pressure of 1,000 psig. The average of the calculated viscosities was 192.0 micropoise with a standard deviation of ± 0.6 micropoise, which gives a 95 per cent confidence limit (twice the standard deviation) of ± 0.6 per cent for the experimental data. The corresponding viscosity given by Michels and Gibson for the same conditions is 191.4 micropoise.

To demonstrate the range of the new instrument, viscosity of water was measured at 1,150 psig at room temperature. The results agreed with the data in the literature¹⁴ within ± 0.2 per cent for 14 of the 15 tests made, and within 0.7 per cent for the other.

With the instrument's accuracy demonstrated over a range of viscosities from 180 (nitrogen) to 9,200 micropoise (water), tests were initiated with propane and ethane. The results are summarized in Table 3. The experimental measurements were made at room temperature, which varied from 24.6 to 26.0°C (76.4 to 78.8°F), and the calculated viscosities were cor-

rected to 25°C (77°F) by use of the data of Smith and Brown.¹³ There is very good agreement between the extrapolated gas phase atmospheric pressure viscosity and the values available in the literature.² The new liquid-phase viscosity data and the Smith and Brown data for propane are plotted in Fig. 9. The data differ by approximately 2 per cent, well within the ± 5 per cent possible error reported by Smith and Brown.

Agreement between the two sets of data in Fig. 9 is very good since they were obtained by two completely different experimental techniques. Smith and Brown in their studies used a rolling-ball viscometer which had to be calibrated with fluids of known viscosity and in the same viscosity and density range as the test fluids. The other instrument was an absolute capillary-tube

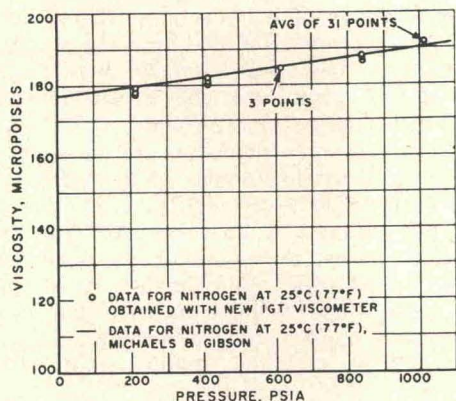


FIG. 8—COMPARISON OF NEW VISCOMETER DATA AND LITERATURE DATA FOR NITROGEN.

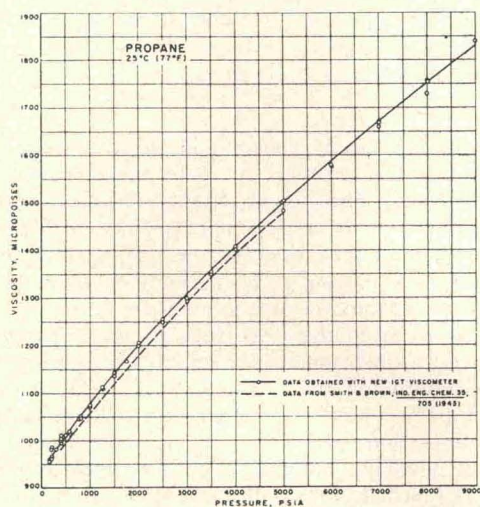


FIG. 9—COMPARISON OF NEW VISCOMETER DATA AND LITERATURE DATA FOR PROPANE.

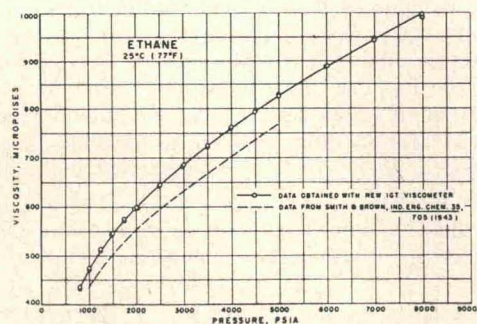


FIG. 10—COMPARISON OF NEW VISCOMETER AND LITERATURE DATA FOR ETHANE.

TABLE 3—EXPERIMENTAL VISCOSITY DATA OBTAINED WITH THE NEW VISCOMETER FOR PROPANE AND ETHANE AT 25°C (77°F)

Pressure (psia)	Propane viscosity (micropoise)	Ethane viscosity (micropoise)
101	82.2 (v)	—
119	82.6 (v)	—
160	957	—
180	965	—
200	970	94.4 (v)
300	981	—
400	1,004	100.8 (v)
500	1,012	106.2 (v)
533	—	110.1 (v)
600	1,014	—
800	1,049	433.5
1,000	1,075	475.0
1,250	1,111	510.7
1,500	1,139	544.6
1,750	1,168	572.8
2,000	1,204	597.6
2,500	1,252	644.5
3,000	1,297	684.3
3,500	1,352	722.2
4,000	1,406	760.0
5,000	1,483	826.1
6,000	1,578	887.2
7,000	1,664	943.4
8,000	1,754	994.2
9,000	1,840	—

(v) = Vapor phase. All other values are for liquid phase.

viscometer, with the viscosity calculated directly from the dimensions of the instrument and flow measurements on the test fluid. That such close agreement exists attests to care and precision of the experimental work by Smith and Brown.

The results for ethane are plotted in Fig. 10, with the ethane viscosity data of Smith and Brown¹³ for comparison. Deviations of up to 8 per cent exist between the two sets of data, which is not too unexpected since the viscosity and density relationships for ethane preclude operation of the rolling-ball viscometer under the required conditions of laminar flow. The rolling-ball viscometer was calibrated under conditions of laminar flow, and the ethane measurements were apparently made in the region of turbulent flow. For these reasons it is believed the new viscosity data presented represent an improvement over data previously available.

SUMMARY

An apparatus has been developed for measurement of viscosity of fluids over the range of reservoir pressures and temperatures. Equations have been derived for calculating viscosity from measurement on isothermal, laminar flow of either compressible or incompressible fluids. Viscosity values calculated from experimental measurements on nitrogen and water give excellent agreement with accepted literature data. Viscosity-pressure data obtained for ethane and propane at room temperature indicated significant deviation from existing literature values. It is concluded that an accurate new research tool has been developed for the study of PVT behavior of natural gas, condensate and high-gravity dissolved-gas systems.

NOMENCLATURE*

- D = diameter
- g_c = dimensionless constant
- N = moles of gas
- P = pressure
- Q = volumetric flow rate
- \bar{u} = average velocity
- v = volume

*See AIME Symbols List in *Trans. AIME* (1956) 207, 363, for other symbol definitions.

V_f = free receiver volume above the electrodes
 z = compressibility factor
 ϵ = electrode spacing
 θ = time

SUBSCRIPTS:

a = average
 D = displaced
 f = test fluid
 lm = log-mean
 m = mercury
 t = transition
 1 = initial condition
 2 = final condition

ACKNOWLEDGMENTS

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REFERENCES

1. Bicher, L. B., Jr. and Katz, D. L.: "Viscosities of the Methane-Propane System", *Ind. and Engr. Chem.* (1943) **35**, 754.
2. Carr, N. L.: "Viscosities of Natural Gas Components and Mixtures." *Bull.*, Institute of Gas Technology (June, 1953) No. 23.
3. Carr, N. L., Kobayashi, R. and Burrows, D. B.: "Viscosity of Hydrocarbon Gases Under Pressure", *Trans. AIME* (1954) **201**, 264.
4. Carr, N. L., Parent, J. D. and Peck, R. E.: "Viscosity of Gases and Gas Mixtures at High Pressure", *Chem. Engr. Prog. Symposium Series* (1955) **51**, No. 16, 91.
5. Comings, E. W., Mayland, B. J., and Egly, R. S.: "The Viscosity of Gases at High Pressures", *Bull. U. of Illinois Engr. Exp. Sta.* (1944) 354.
6. Ellington, R. T. and Eakin, B. E.: "A Summary of Methods for Measuring Fluid Viscosities and Compressibilities at High Pressures", *Proc. National Conf. on Industrial Hydraulics* (1957) **11**, 163.
7. Fisher, W. J.: "The Coefficients of Gas Viscosity—II", *Phys. Rev.* (1909) **28**, 73.
8. Golubev, I. F.: "Viscosity of Gases and Gas Mixtures at High Pressure", *Jour. Tech. Phys.*, USSR (1938) **8**, 1932.
9. Langhaar, H. L.: "Steady Flow in the Transition Length of a Straight Tube", *Trans. ASME* (1942) **64**, A-55.
10. Michels, A. and Gibson, R. O.: "The Measurement of the Viscosity of Gases at High Pressures—The Viscosity of Nitrogen to 1000 Atmospheres", *Proc., Roy. Soc.* (1931) **134A**, 288.
11. Rankine, A. O.: "A Simple Viscometer for Gases", *Jour. Sci. Instruments* (1924) **1**, 105.
12. Sage, B. H. and Lacey, W. N.: "Effect of Pressure Upon Viscosity of Methane and Two Natural Gases", *Trans. AIME* (1938) **127**, 118.
13. Smith, A. S. and Brown, G. G.: "Correlating Fluid Viscosity", *Ind. and Engr. Chem.* (1943) **35**, 705.
14. Swindells, J. F., Coe, F. R., Jr. and Godfrey, T. B.: "Absolute Viscosity of Water at 20° C", *Jour. Res.* (1952) **48**, 1.
15. Walters, R. and Eakin, B. E.: "Simplified High-Pressure Electrode Assembly", *Rev. Sci. Instr.* (1957) **28**, 204.

APPENDIX

THEORY OF FLUID FLOW IN THE VISCOMETER

The equation for compressible, steady, isothermal, laminar flow in a capillary,

$$P_1 - P_2 = \frac{2(z_2/z_1)}{(1 + P_2/P_1)} \frac{32\mu\bar{u}L_c}{gD_c^2}, \dots (2)$$

reduces to the Hagan-Poiseuille equation for incompressible flow,

$$P_1 - P_2 = \frac{32\mu\bar{u}L_c}{gD_c^2} \dots (3)$$

even for gases if operating pressure is substantially above atmospheric. Further consideration of the flow problem required treatment of the pressure drop resulting from the flow of mercury through the composite channel between reservoir and receiver and so-called "end effects". These are normally considered losses due to sudden expansions and contractions in flow channel, such as at the ends of tubes.

The acceleration of a fluid from rest to steady viscous flow requires a certain distance of travel for establishment of final velocity distribution. Until this point is reached the resistance to flow is greater than predicted by Poiseuille's law, as was shown by Langhaar.⁹ By applying a linearizing approximation to the Navier-Stokes equation and defining the transition length, L_t , as the distance required for the centerline fluid velocity to reach 99 per cent of its final value, Langhaar obtained

$$L_t = 0.23r_c(2r_c\bar{u}\rho_f)/\mu \dots (4)$$

For tubes with lengths much greater than L_t , the fundamental equation for the tube-type viscometer becomes,

$$-\Delta P = \frac{8\mu_f Q L_c}{g\pi r_c^4} + \frac{\beta Q^2 \rho_f}{g\pi^2 r_c^4} \dots (5)$$

in which β is nearly constant at the theoretical value of 1.14 with experimentally determined values from 1.11 to 1.18.¹⁴ The kinetic energy term accounts for full development of a parabolic velocity profile. When true end effects are important the value of β depends upon the flow pattern near the ends of the tubes and the transition from one size to another.

Transition from the large tube to the flow capillary occurs in this instrument in a smooth converging nozzle with a contraction ratio of about 15:1, and at the end of the tube in a similar diverging nozzle. The expansion and contraction processes exactly compensate each other in adiabatic compressible flow, and very nearly so in isothermal, incompressible flow. It is therefore believed that these end effects are negligible for this design.

It is recognized that in this viscometer, some head loss will result from passage of each of the fluids through the elbows in the flow paths. Every effort was made to minimize these losses by providing as few changes in direction as possible and making the flow channels through them as large as possible. Losses in head due to these effects were neglected in developing correction terms.

The final equation representing steady-state flow for the modified Rankine viscometer can be written as follows.

$$-\Delta P = \frac{8\mu_f Q_f L_c}{g\pi r_c^4} + \frac{\beta Q_f^2 \rho_f}{g\pi^2 r_c^4} + \frac{8\mu_m Q_m L_m}{g\pi r_m^4}, \dots (6)$$

where ΔP is the indicated pressure differential due to the difference in mercury levels in the mercury reservoir and receiver, given by

$$\Delta P = \frac{g(\Delta h)(\rho_m - \rho_f)}{g_c} \quad (7)$$

If the cross-sectional areas of the two vessels are equal,

$$-A_1 \frac{dh_1}{d\theta} = A_2 \frac{dh_2}{d\theta} \quad (8)$$

or, the decrease in mercury level in the reservoir is equal to the increase in level in the receiver. This means that

$$\frac{d(\Delta h)}{d\theta} = 2 \frac{dh_1}{d\theta} = -2 \frac{dh_2}{d\theta} \quad (9)$$

and that the difference between initial and final mercury differential will be equal to twice the electrode spacing,

$$\Delta h_2 - \Delta h_1 = -2\epsilon \quad (10)$$

At any given time, θ , the general equation representing steady flow in the viscometer can be written,

$$\Delta P = a(\Delta h) = b_f Q_f + c_f Q_f^2 + b_m Q_m \quad (11)$$

where $a = (\rho_m - \rho_f)g/g_c$, $b = 8\mu L/g\pi r^4$, and $c = \beta\rho/g\pi^2 r^4$.

Q is instantaneous flow rate at time, θ , and subscripts f and m refer to the fluid and mercury, respectively.

The instantaneous mercury flow rate, Q_m , can be expressed as a function of the area of the receiver, A , and the rate of change of h_2 , $dh_2/d\theta$, as

$$Q_m = A \frac{dh_2}{d\theta} = -\frac{A}{2} \frac{d(\Delta h)}{d\theta} \quad (12)$$

If the fluid being displaced is incompressible,

$$Q_m = Q_f = -\frac{A}{2} \frac{d(\Delta h)}{d\theta}$$

The assumption of an incompressible test fluid is valid at this point in the derivation, since only differential changes in mercury level are being considered.

The differential equation for the viscometer now becomes,

$$-a(\Delta h) = (b_f + b_m) \frac{A}{2} \frac{d(\Delta h)}{d\theta} + c_f \frac{A}{2} \left[\frac{d(\Delta h)}{d\theta} \right]^2 \quad (13)$$

Integration of the roots of Eq. 13 results in two equations, one of which gives a negative flow time and was therefore discarded. The second solution, with integration between the limits of time = 0 and θ , and $\Delta h = \Delta h_1$ and Δh_2 ,

$$\theta = -\frac{(b_f + b_m)}{2c_f} \left\{ \ln \frac{\Delta h_2}{\Delta h_1} + \alpha_2 - \alpha_1 + \ln \frac{(\alpha_2 - 1)(\alpha_1 + 1)}{(\alpha_2 + 1)(\alpha_1 - 1)} \right\} \quad (14)$$

where

$$\alpha_1 = \sqrt{1 + \frac{8ac_f \Delta h_1}{A(b_f + b_m)^2}}$$

$$\alpha_2 = \sqrt{1 + \frac{8ac_f \Delta h_2}{A(b_f + b_m)^2}}$$

This rigorous solution of the flow equation for the

viscometer requires use of an iterative procedure to solve for the fluid viscosity.

To obtain a first approximation of the fluid viscosity to use in solving Eq. 14, Eq. 13 was solved assuming Langhaar's correction term to be constant and applicable to the integrated form of the modified equation. This resulted in an equation which may be solved directly for viscosity.

$$\mu_f = \frac{\pi g r_c^4 \theta}{8 L_c V_D} \left[\frac{\Delta h_{1m} (\rho_m - \rho_f) g}{g_c} - \frac{\beta \rho_f V_D^2}{g \pi r_c^4 \theta^2} \right] - \frac{L_m r_c^4 \mu_m}{r_m^4 L_c} \quad (15)$$

which has been termed a "pseudo-steady-state" equation. Both equations were programmed for solution by use of IGT's electronic digital computer, ALWAC III, and the calculated viscosity values for the two equations agree within 0.1 per cent. By using the viscosity value obtained by solution of the pseudo-steady-state equation as the first approximation in the rigorous equation, very rapid convergence is obtained.

EFFECT OF COMPRESSIBILITY ON VOLUME OF FLUID DISPLACED THROUGH THE CAPILLARY TUBE

For an incompressible fluid the volume contained in the receiver between the electrodes is the actual volume displaced through the capillary tube. However, since the pressure in the mercury receiver decreases slightly during the run, the fluid confined in the receiver and fittings will expand, resulting in a slightly larger volume being displaced through the capillary tube than the volume of mercury contained between the electrodes. The correction is derived as follows.

Total moles of gas initially in receiver when mercury level contacts the long electrode,

$$N_1 = \left(P_a + \frac{\Delta h_2}{2} + \epsilon \right) (V_F + V_m) / zRT \quad (16)$$

and moles of gas remaining when mercury contacts the short electrode,

$$N_2 = \left(P_a + \frac{\Delta h_2}{2} \right) V_F / zRT \quad (17)$$

The moles displaced are, therefore,

$$N_D = N_1 - N_2 \quad (18)$$

or

$$P_a V_D = \left(P_a + \frac{\Delta h_2}{2} + \epsilon \right) (V_F + V_m) - \left(P_a + \frac{\Delta h_2}{2} \right) V_F \quad (19)$$

then

$$V_D = V_m \left(1 + \frac{\Delta h_2 + 2\epsilon}{2 P_a} \right) + \frac{\epsilon V_F}{P_a} \quad (20)$$

For liquids, and for gases at pressures above 1,500 psia, the correction was less than 0.1 per cent and was therefore neglected in the calculations and V_m used as the volume displaced through the capillary tube. For gases at pressures less than 1,500 psia the corrected V_D was employed. ★★★