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		Physical Properties of Hydrocarbons	PAGE 1 OF 12	

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SUBJECT: Method for Determination of Liquid Density and Viscosity of Organic Coolants over the Temperature Range (M.P. to 850°F)

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I STATEMENT OF PROBLEM

To measure the liquid density and viscosity of organic coolants over the temperature range 300 to 850°F.

II. SUMMARY

A knowledge of the density and viscosity of organic coolants as a function of temperature and extent of decomposition is important to reactor operation. Techniques were developed to measure these properties over the temperature range 300 to 850°F.

1. Density

Density is determined as a function of temperature by measuring the volume occupied by a known weight of sample in a modified Lipkin pycnometer. The pycnometer is pressurized with an inert gas to prevent boiling and maintained at thermal equilibrium in a hot air furnace. Estimated accuracy of measurements is ± 0.3 per cent.

2. Viscosity

Viscosity is determined as a function of temperature by measuring efflux times in a (Cannon-Manning) modification of the Ostwald semi-micro viscosimeter. The Ostwald pipet is pressurized with an inert gas to prevent boiling. Estimated relative accuracy of measurements is ± 3 per cent.

III. APPARATUS, METHOD, ACCURACY, AND SAMPLE CALCULATIONS

A. Apparatus

The density and viscosity determinations are carried out using the same constant temperature air bath and temperature measuring equipment. For this reason, the apparatus for both is described jointly. A photograph of the assembled apparatus is shown in Figure 1.

The equipment consists essentially of (1) a modified Lipkin pycnometer for determining the density by measuring the volume of a given weight of sample at a given temperature (see A, Figure 2); (2) a cathetometer to measure accurately the volume difference in the pycnometer at various temperatures (see C, Figure 1); (3) a hot air furnace to maintain thermal equilibrium throughout the sample (see B, Figure 1); (4) a means of measuring temperature accurately; (5) a viscosimeter to determine viscosity at a given temperature by measuring the rate of flow through a section of capillary tubing; and (6) an inert gas system to maintain a pressurized inert gas blanket over the sample to prevent boiling.

The pycnometer used to measure density is a 1 ml modified Lipkin pycnometer obtained from the H. S. Martin Company (see A, Figure 2). The two legs have been modified in order to facilitate glass-to-metal connection to the pressurizing system. A 3-in. long section of 14.0 mm O.D. by 8.0 mm I.D. heavy-walled glass tubing is attached to the bulb leg and a 6.4 mm O.D. by 1 mm capillary to the opposite leg to permit mating with 1/2-in. and 1/4-in. Swagelok fittings, respectively.

A standard Gaertner cathetometer is used to measure the height of liquid in the two legs to determine volume.

An Ostwald-Cannon-Manning semi-micro type viscosimeter, size 50, is used to determine viscosity and was obtained from the Cannon Instrument Company. The viscosimeter was modified similarly to the pycnometer (see Figure 1) to provide for the glass-to-metal connection. A 4-in. section of 12.4 mm O.D. x 8.2 mm I.D. thick-walled glass tubing was attached to the large bore leg and 6.4 mm O.D. x 0.5 mm capillary tubing to the other leg.

The hot air furnace for maintaining temperature conditions was designed and fabricated at Atomics International. The furnace is essentially a 16 gauge plate steel box 17-in. high x 14-in. wide x 14-in. deep lined with 1/2-in. thick transite. An exterior wall of firebrick provides the necessary additional thermal insulation. Heat is supplied by four 400 watt tubular heaters mounted on the side walls and a 400 watt tubular heater mounted on the floor. A 3-in. diameter hole, for positioning the pycnometer and viscosimeter in the furnace, is located in the top surface.

The pycnometer and viscosimeter are mounted in a pipette block (see A, Figure 1) which slips over studs on the rim of the 3-in. holes. The pipette block is secured to the furnace frame and proper vertical alignment of the pipettes is assured. Two Pyrex windows 15-in. high x 5-in. wide x 1/2-in. thick are mounted in the front and rear faces of the furnace. The front window permits observation of the fluids in the pycnometer and viscosimeter. A strong light with reflector is placed at the rear window to provide adequate illumination of the fluids in the two instruments. The temperature in the furnace is manually regulated by means of Powerstat autotransformer controls. The general temperature level is maintained by the side wall heaters controlled by a single 20 amp. Powerstat mounted on the lower right side panel not visible in Figure 1. The vertical temperature gradient is controlled through a small Variac regulating the output of the floor heater unit. The temperature gradient over the pycnometer and viscosimeter sections containing fluid is about 8°F at 800°F.

The pressure system consists of a helium gas cylinder, pressure regulating valve, and a manifold fabricated from 1/4-in. copper tubing which is used for manipulation of pycnometer and viscosimeter liquid levels. A schematic diagram of the pressure system is shown in Figure 3. Pressure is measured by a calibrated pressure gauge (G). Liquid levels in the viscosimeter are manipulated in the legs by means of Valves V-7 and V-5. By closing V-7 and opening V-5, pressure on the V-2 leg is decreased and the liquid raised above the upper etched line on the bulb leg of the viscosimeter. By closing V-5 and opening V-7, the pressure is equalized in both legs and the liquid flows downward by gravity and the efflux time is measured as the interface moves from the upper to lower etched lines on the viscosimeter.

The temperature is measured by means of three chromel-alumel thermocouples located so as to measure the temperature at approximately the lower, upper, and midpoints of liquid columns in the two instruments. A Rubicon B potentiometer (see B, Figure 1) is used to measure the e.m.f. of the thermocouples. The thermocouples are calibrated with N.B.S. platinum-rhodium thermocouples.

B. Method

1. Density

The weight of the sample may be determined accurately by weighing the pycnometer before and after it is filled. The volume of the sample, however, must be established from a calibration of the pycnometer volume with a fluid of known density.

Calibration of the pycnometer is accomplished by filling the pycnometer with triple distilled mercury to an etched line (zero point) just above the bulb and determining the weight of mercury. From the weight and density, the volume of the pycnometer up to this zero point is established. Increments of mercury are added and the height of mercury in the legs with reference to this zero point is measured with the cathetometer. The liquid height in the legs is then related to the volume which is calculated from the weight of mercury and its density. The calibration consists essentially of a plot of the volume of the pycnometer as a function of the height of liquid above the zero point.

Loading of the pycnometer with organic coolant sample is accomplished by filling a 5 cc syringe with hydrocarbon and injecting the sample into the capillary of the leg opposite the bulb leg. The pycnometer is supported vertically in a semi-circular heater which maintains a temperature of 20 to 30°F above the melting point. The 5 cc syringe filled with sample is maintained above the melting point by directing the flow of hot air from the heat gun toward it. The No. 20 hypodermic stainless steel needle is inserted into the capillary, which has been tapered to assure a tight fit. A continuous stream of sample is forced down the capillary and into the bulb. About 0.7 ± 0.1 cc are introduced in this manner. Introduction of too large sample will result in high liquid interface at high temperatures which cannot be observed. Too small a sample will result in failure of the liquid to rise up to the capillary section at low temperatures (400-500°F) thus preventing an accurate volume measurement. The pycnometer is weighed before and after loading and weight of sample is determined by difference.

Next, a 1/4-in. Swagelok nut is slipped over the small end of the pycnometer, followed by a nylon ferrule, similar to Item 2, Figure 3, with the large diameter facing the nut. Last, a nylon ferrule, similar to Item 1, Figure 2, is placed on the tube with the large diameter facing the nut. The assembly is then placed in the pipette block and the Swagelok connection carefully tightened so as not to over-stress the glass. Similarly, the large end of the pycnometer is fastened with a 1/2-in. Swagelok connection to the end of the copper tube loop projecting from the top of the pipette block. Care must be exercised in aligning the loop end before the connection is tightened in order to avoid introduction of strains in the pycnometer tube.

The system is alternately evacuated and flushed four times with helium by manipulating valves marked vac. and PRV. The system is finally pressurized to 50 psi and the furnace is brought up to temperature. The lower heater is adjusted to stabilize the temperature.

The cathetometer is leveled and a reading is taken of the zero mark after the furnace and pycnometer are at thermal equilibrium. The height of the liquid interface above the zero mark is measured with the cathetometer. From the mercury volume calibration, the volume of sample is determined. The calculation of density follows from definition.

2. Viscosity

The determination of viscosity involves measuring the rate of flow of a given volume of liquid through a capillary orifice. By calibrating the viscosimeter with a solution of known viscosity, the relative viscosity of unknown material may be determined. The following relationship of the viscosity of a Newtonian liquid in laminar flow within a capillary has been derived from Poiseuille's equation:

$$\eta = C\rho t - B\rho t^{(1)}$$

where η = viscosity in centipoise

ρ = density in g/cc

t = time of flow in seconds

C = a constant established by calibration of the viscosimeter with a fluid of known viscosity

$$B = \frac{mv}{81\eta t^2}$$

where m = constant, usually assumed to be one

By careful design of the pycnometer, the "B" term may be reduced to a negligible value. For our work, this term is neglected and the equation is reduced to the general form,

$$\eta = C\rho t$$

The constant C is a property of the particular viscosimeter and is evaluated using distilled water as a calibrating fluid.

Calibration of the viscosimeter is accomplished by introducing about 2 cc of distilled water. The weight of water is determined accurately by weighing the viscosimeter before and after loading. The viscosimeter is mounted in a vertical position in a constant temperature water bath and is allowed to come to thermal equilibrium. A rubber hand suction pump is placed over the large tube opening and the water interface in the opposite leg is gently raised above the upper etched line located just above the bulb in the capillary. The bulb is removed and time required for

the interface to pass from the upper line to the lower etched line located just below the bulb is measured to the nearest 0.1 second. Three such determinations are made and should not differ by more than 0.1 seconds. If results are not in close agreement, the viscosimeter is not clean or the water is contaminated. Knowing viscosity, density, and efflux time, "t", the constant "C" can be calculated. The "C" so obtained must be corrected when different sample volumes and temperatures are used from those in the calibration. The correction is given below in the last paragraph of this section.

The loading of the viscosimeter is done in a manner similar to that for loading the pycnometer. A 2 cc graduated pipette is used instead of syringe to transfer the sample. The sample is loaded through the large diameter tube instead of the capillary as in the case of the pycnometer. The length of the pipette permits placing the sample in the bottom of the leg without spreading it over the sides. The viscosimeter is connected to the pipette block in the same manner as the pycnometer. Normally, the viscosimeter and pycnometer are mounted in the furnace so that both determinations may be made at the same temperature.

See the procedure under density for removing air and covering samples with an inert gas blanket. The raising and lowering of the interface in the bulb leg is accomplished by closing Valve V-7 and opening V-5. This results in maintaining pressure on the large diameter leg and decreasing pressure slightly on the bulb leg as the inert gas escapes through the pen hole. The differential pressure causes the interface to rise in the bulb leg. When the desired interface height is reached, Valve V-5 is closed and V-7 opened. Pressure equalizes across both legs and gravity causes the fluid flow. The interface is timed as it traverses the two etched lines and the efflux time is obtained for the sample. Repeat three times and use the average time to calculate viscosity from,

$$\eta_2 = C_1 \left(1 - \frac{V_2 - V_1}{0.785 H d^2} \right) \rho_2 t_2$$

where η = viscosity of hydrocarbon, centipoise

C_1 = constant derived from water calibration

V_2 = volume of sample, cm^3

V_1 = volume of water in calibration

ρ_2 = density of hydrocarbon, g/cm^3

t_2 = efflux time of hydrocarbon, seconds

H = driving liquid head, cm

d = working diameter of lower reservoir, cm

The term, $1 - \frac{V_2 - V_1}{0.785 H d^2}$, is essentially a correction for variations in pressure head due to volume differences arising between sample at temperature of measurement and volume of calibration sample at room temperature⁽²⁾.

3. Cleaning Procedure

The pipette is filled with dibutyl phthalate and heated in the half shell heater used for loading. The deposits are softened by the action of heat and phthalate. The residue is flushed out with a trisodium phosphate cleaning solution. The pipette is then heated, cooled, and flushed with distilled water. The procedure is repeated if necessary until the pipettes are clean. The pipette is dried in an oven at 120°C before loading.

C. Accuracy

1. Density

The weight of sample and volume can be determined accurately to the fourth place. The average temperature is not known to better than 0.3 per cent. The estimated obtainable accuracy is ± 0.1 per cent, however, the precision obtained is about ± 0.2 per cent. The absolute accuracy is estimated to be about 0.3 per cent.

2. Viscosity

The weight and volume of sample can be determined to the fourth place. The temperature is known to ± 0.3 per cent. The constant may be evaluated to about 1 per cent. The precision obtained is approximately 3 per cent. The relative viscosity is estimated to be accurate to about 3 per cent.

D. Sample Calculations

1. Density

1. Temperature = 500°F
2. Reading left leg = 73.71
3. Reading right leg = 73.71
4. Zero-point = 73.17
5. Zero-points volume = 0.7684
6. Weight of sample

pycnometer + sample weight = 77.4112

pycnometer + sample weight = 76.6747

sample weight = 0.7365

7. Determine volume of liquid in capillary

Right leg	73.71 cm	Left leg	73.71 cm
	<u>-73.17</u>		<u>-73.17</u>
	0.54 cm		0.54 cm

Δ Volume = 0.0068 from mercury calibration for leg height

Zero point volume = 0.7684

Total volume = 0.07684 + 0.0068

8. Density

$$= \frac{0.7365}{0.7684 + 0.0068}$$
$$= 0.950 \text{ g/cc}$$

2. Viscosity

1. Calibration constant evaluated at room temperature (known to nearest 0.1°F) with water, C = 0.002790
2. Volume of water used in calibration = 2.2046 cc
3. Temperature of organic coolant sample = 500°F
4. Efflux times for organic coolant at 500°F

Trial 1. t = 348.4 sec.
Trial 2. t = 348.6 sec.
Trial 3. t = 348.5 sec.
Average t = 348.5 sec.

5. Density of organic sample at 500°F = 0.9510 g/cc, experimentally determined.

6. Volume of sample

Viscosimeter + sample weight = 106.9276 g
Viscosimeter weight = 104.9724
Weight of sample = 1.9552 g

$$\text{Volume} = \frac{1.9552}{0.9510} = 2.0559 \text{ cc at } 500^\circ\text{F}$$

7. Average driving head at 500°F, H = 10.1 cm
8. Reservoir diameter, d = 0.8 cm

9. Viscosity at 500°F

$$\begin{aligned}\eta &= C \left[1 - \frac{(V_2 - V_1)}{(0.785H d^2)} \right] t_r \\ &= 0.002790 \left[1 - \frac{(2.0559 - 2.2046)}{(0.785)(10.1)(0.8)^2} \right] (348.5)(0.9510) \\ &= 0.952 \text{ centipoise}\end{aligned}$$

IV. REFERENCES

- (1) Weissberger, A., Physical Methods of Organic Chemistry, pg. 334, Vol. 1, Part 1, (1949), Interscience Publishers, Inc., New York
- (2) Cannon, M.R. and Fenske, M.R., Industrial and Engineering Chemistry, Analytical Edition, 10, 291 (1938)

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FIGURE 1

FIGURE 2

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MODEL NO.

FIGURE 3
SCHEMATIC OF PRESSURE
SYSTEM

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