

REVIEW

## A Review on Viscometer Devices for Gas Phase

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**Abstract** -Viscosity, one of the transport properties, is a basic property required for developing any device in which fluid flows. In the present paper, major gas viscometers devices are reviewed. The review is covering to the pressures and the temperatures ranges, methods and accuracy of the instruments, and the coefficient of viscosity-range of the sample. This study is expected become useful information for any researcher who wants to develop a gas viscometer apparatus in a near future. By identifying the coefficient of viscosity-range and the measurement-range of temperatures and pressures, then we can choose the effective and efficient method to be developed.

**Keywords:** Capillary, falling body, oscillating disk, rotating cylinder, and, vibrating wire.

### Introduction

The molecular transfer process in fluids, internal friction/viscosity, thermal conductivity and thermal diffusivity are very important for explaining physical and physicochemical phenomena, and also for solving many chemical-engineering problems involving mass, momentum and heat transfer. A viscometer is an instrument used to measure the viscosity of a fluid. Viscosity, one of the transport properties, is a basic property required for developing any device in which fluids flows. In the present paper, major gas viscometers devices are reviewed. The review is regarding to the pressures and the temperatures ranges, coefficient viscosity-range, method of the measurement and sample of the gas. This study is expected become useful information for any researcher who wants to develop a viscometer apparatus in a near future. By identifying the coefficient of viscosity-range of the sample and the measurement range of temperatures and pressures, then we can choose the effective and efficient method to be developed. There are five major viscometer instruments, which is used to measure gas viscosity, and already established and published in a prominent journal. Falling-body, Capillary tube, Rotating cylinder and Oscillating Disk, are the methods involve hydrodynamic correction and approximations for the end, edges and walls. The corrections are often large and sometimes are the primary source of error. Vibrating wire methods is the last major methods that can eliminate hydrodynamic correction term (Tough *et al.*, 1964). Those are will describe in detail in the section of the present paper.

### Literature Survey on Viscometer Devices

Literature survey has been done related on five major viscometer devices for gas phase. The instrument which is has been published in a prominent journal will describe in detail in this section and tabulated in Table 1.

#### Falling-body method

Falling-body (FB) viscometer involves the use of the time of free-fall of an object under the influence of gravity through the sample fluid. Generally, falling body is a body revolution and has most commonly been a sphere or cylinder shape. The instruments are not among those of the highest accuracy because it is rather difficult to ensure that the instrument operates in line with a theory of it. However, exceptionally, such instruments have been used for accurate measurements including wok in Japan on a new standard reference viscometer (Wakeham *et al.*, 1991). An alternative realization of a falling-body viscometer makes use of a right circular cylinder falling under the influence of gravity along the axis of a coaxial cylindrical tube. This arrangement has the advantages that it is possible to secure a low Reynolds number for the fluid flow by choosing a suitable geometry, or by reducing the cylinder's mass by making it hollow. A schematic diagram of Rudenko's apparatus based on falling body method is shown in Figure 1 (Rudenko and Slyusar, 1968). A duralumin tube ( $L_T = 190$  mm,  $OD = 7$  mm and  $r_2 = 1.51$  mm) was inserted inside the high pressure bomb ( $L = 255$  mm,  $OD = 30$  mm,  $ID = 7.5$  mm made of beryllium bronze). A duralumin cylinder ( $L_s = 12.5$  mm,  $r_1 = 1.505$  mm) inside, which is pressed a column of magnetic alloy, falls in the tube. For a cylinder of radius,  $r_1$  and length,  $L_s$ , in a tube of radius,  $r_2$  and length,  $L_T$ , the viscosity is given by

$$\eta = \frac{(1 - \rho / \rho_m) t}{A} \quad (1)$$

Where

$$A = \frac{2\pi L_s L_T}{mg \left\{ \ln(r_2 / r_1) - \left[ (r_2^2 - r_1^2) / (r_2^2 + r_1^2) \right] \right\}} \quad (2)$$

Here  $\rho$  is the density of the sample fluid and  $\rho_m$  is the density of the material of the cylinder, while  $L_T$  is the distance travelled by the falling body of mass  $m$  in a gravity of  $g$  and  $t$  is the traveling time. Rudenko and Slyusar

(1968) have measured hydrogen viscosity for pressure up to 217 MPa and temperature up to 300 K with this apparatus.

The traveling time of the cylinder was determined with a chronometer (0.1 sec of resolution) by reflections of a galvanometer mirror caused by passage of the cylinder through the inductive coils which are connected in opposition in order to compensated for the earth's magnetic field when inverting the viscometer. The error in measuring the viscosity with this instrument fluctuated from 4 % at high density to 6 % at low density.

**Capillary tube method**

The principle of the capillary tube (CT) method is based on the Hagen-Poiseuille equation of fluid dynamic which was first formulated on the basis of experiments by Hagen in 1839 and Poiseuille between 1840 to 1846 (Wakeham *et al.*, 1991). When a sample gas flows through a thin capillary tube with a radius and a length of  $a$  and  $l$ , respectively. For a certain volumetric flowrate,  $Q$ , then a pressure drop,  $\Delta P$ , can be obtained. Since two pressure taps are required for measurement of the pressure drop, the velocity distribution can be realized only some distance downstream from the inlet of the capillary. Even before flowing into the tube, the flow must converge and accelerate since diameter of the chamber is much larger than that of the capillary. Furthermore, at exit, the flow from the tube creates a jet into the exit chamber. Both of these effects contribute to the measured pressure drop. Figure 2 shows a capillary tube viscometer used by Yusibani *et al.* (2011) to measure hydrogen at high pressure and temperature. Rigorous analyses require term for the so-called kinetic energy correction and the edge correction to be added to the theory as expressed in Eq. 3.

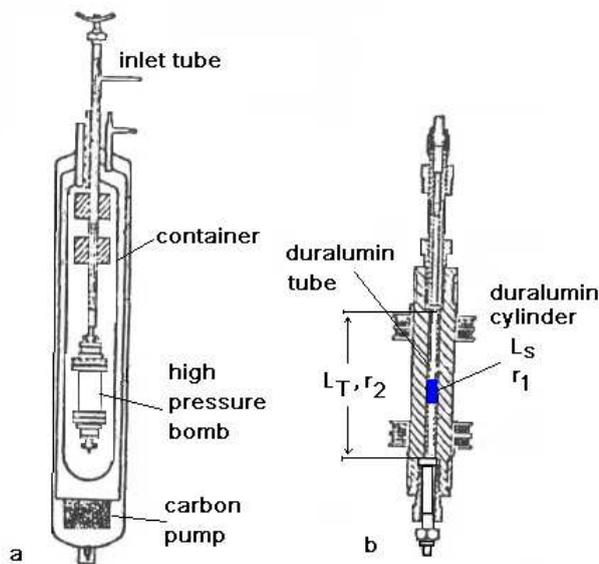
$$\eta = \frac{\Delta P \pi a^4}{8Q(l + na)} - m \frac{\rho Q}{8\pi(l + na)} \tag{3}$$

where  $\rho$  is the density,  $a$  is the inner diameter of the capillary,  $l$  is the length of the capillary,  $n$  is the edge-correction factor of the capillary length and  $m$  is the kinetic-energy correction factor. The above equation is derived on the assumption that: The capillary is straight with a uniform circular cross section, the fluid is incompressible and Newtonian, the flow is a laminar and there is no slip at the capillary wall. When a compressible fluid flows through the capillary, the density of the fluid changes along the capillary in response to the pressure drop. The Hagenbach-Poiseuille equation (Eq. 3) therefore has to be modified including this changes, since the original equation assumed an incompressible fluid. The modified equation is expressed as (Wakeham *et al.*, 1991)

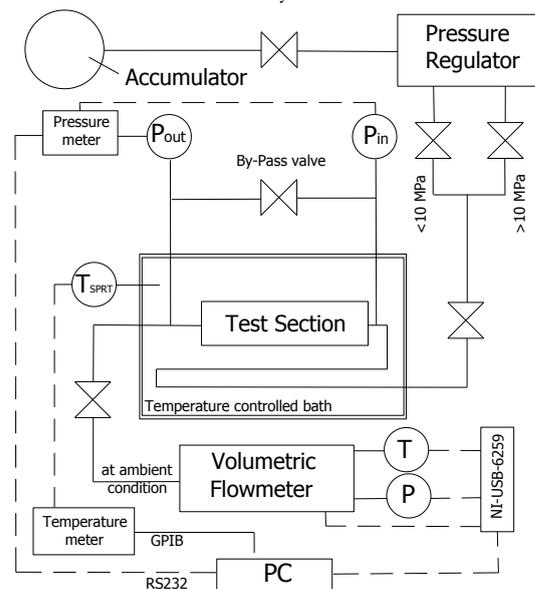
$$\eta = \frac{\pi a^4 (P_{in} - P_{out})}{8Q_m (l + na)} \frac{P_{in} + P_{out}}{2P_m} - \frac{\rho_{in} Q_{in}}{8\pi(l + na)} \left( m + \ln \frac{P_{in}}{P_{out}} \right) \tag{4}$$

This equation approaches Eq. 3 at high pressure. Even at low pressure, however, we can still use Eq. 3 by substituting the average density along the capillary tube. This density correction leads to the same viscosity calculation value as that evaluated from Eq. 3 (Yusibani *et al.*, 2011).

The most difficult measurement in this method is the absolute determination of the diameter of the capillary tube since the radius rises to the fourth power in the viscosity calculation. For measurements of gases at high pressure, the measurement of the flowrate is also difficult and correction for compressibility has to be considered. The pressure drop between the two ends of a capillary is measured most often by mercury U-tube. However, it is not convenient to use mercury in a such of extreme condition with a flameable gas especially in the rural area. The measurement of a pressure drop with a high accuracy at extreme condition is also one of the challenges for this method. Furthermore, the differential pressure sensor at high pressure up to 100 MPa is not available commercially.



**Figure 1.** Falling-body method apparatus used by Rudenko *et al.* (1968) (a) general view of viscometer (b) vertical section of bomb



**Figure 2.** Capillary tube apparatus used by Yusibani *et al.* (2011)

**Table 1.** Viscometer instruments

References	Method	Sample	$\eta$ , uPa.s range	P, (MPa), T, (K) range	Accuracy (%)
<i>Falling body method</i>					
Dindar <i>et al.</i> (2002), Rudenko and Slyusar (1968)	High-pressure falling body viscometer	n-pentane, carbon-dioxide, hydrogen	1.7 - 300	P=10 to 45, 214 (at 16.6 to 300 K) T=323 to 398	Up to 3.8
<i>Capillary method</i>					
Yusibani <i>et al.</i> (2011), Tsederberg <i>et al.</i> (1965), Guevara <i>et al.</i> (1969)	Conversional capillary tube viscometer	Hydrogen, helium, nitrogen, Argon, Air	8 to 195	P= 0.1 to 100 T=295 to 400 (at 100MPa), 900(at 50 MPa), 2150 (at 0.1 MPa)	0.1 to 3
Hilton and Sciver (2007)	A pressurized Gravitational Capillary	Subcooled liquefied gases (oxygen, nitrogen, hydrogen and methane)	~195	P=0.15 to 1 T=Normal boiling to near the freezing point	1 (k=2)
May <i>et al.</i> (2006), May <i>et al.</i> (2007)	Two-capillary viscometer	Argon, hydrogen, CH <sub>4</sub> , Xe	8-28	P=0.1 T=200-400	0.02 up to 0.09
Shin <i>et al.</i> (2001)	Mass-detecting capillary viscometer	water	1000	Ambient condition	ns
Michels and Gibson (1932)	Transpiration method	Nitrogen, hydrogen, deuterium	8 to 52	P=up to 200 T=298 to 398	0.1 to 0.5
<i>Rotating cylinder method</i>					
Daridon <i>et al.</i> (2011), Mason (1947), Diller (1965)	Quartz Crystal Resonators	Parahydrogen, Heptanes, toluene, n-decane with dissolved methane	8 to 984	P= up to 80 T=293 to 332	0.5 to 5
Hurly <i>et al.</i> (2003)	Green span viscometer	Ar, CH <sub>4</sub> , C <sub>3</sub> H <sub>8</sub> , N <sub>2</sub> , SF <sub>6</sub> , CF <sub>4</sub> , C <sub>2</sub> F <sub>6</sub>	8-27	P= up to 3.3 T=210 to 375	0.55 and 1.9 at high density
Evers <i>et al.</i> (2002)	Compact Viscometer-densimeter	Nitrogen, Methane, Helium, neon, Argon, krypton	8-39	P=up to 30 T=233 to 523	Up to 1
<i>Oscillating body method</i>					
Kestin and Leidenfrost (1959), DiPippo <i>et al.</i> (1967), Nabizadeh and Mayinger (1999), Nabizadeh and Mayinger (1989), Johnston and McCloskey (1939), Seibt <i>et al.</i> (2009), Dahestru <i>et al.</i> (2011)	Oscillating disk viscometer	Air, oxygen, nitrogen, nitric oxide, nitrous oxide, carbon dioxide, methane, Hydrogen, H <sub>2</sub> +natural gas, Helium, Ne-N <sub>2</sub> , He-CO <sub>2</sub> , N <sub>2</sub> -Ar, He-Ar, He-Ne, Ne-Ar, Ar-NH <sub>3</sub> , Refrigerants (R12, R113, R114, R12+R114, R404A, R407C, R410A, R507)	8 to 31	P=0.1-13.7 T=90 – 510	0.2 at low T, 0.5 at high T, 1 for refrigerant along saturation line
<i>Vibrating method</i>					
Wakeham <i>et al.</i> (1991), Charles (1983), Black <i>et al.</i> (1971), Kestin <i>et al.</i> (1959), Kestin and Whitelaw (1963), Restina <i>et al.</i> (1987), Wilhem <i>et al.</i> (1998)	Straight vibrating wire viscometer	Helium, nitrogen, toluene, Octane, Heptane, Decane, Argon, Methane, Squalane, N10, S20, S60	8 to 10 <sup>6</sup>	P= 0.06 to 166 T=283 to 473	0.3 to 10
Trappeniers <i>et al.</i> (1980), Van der Gulik <i>et al.</i> (1988)	Curved vibrating wire viscometer	Liquid helium 3	20 to 1000	P=0.01 to 2.9 T=0.006 to 3	ns

Ns= not specified

**Torsional crystal method**

The torsional-crystal (TC) method was introduced by Mason *et al.* (1947). If cylinder of piezoelectric material is cut with its axis along the *x*-axis, and a sinusoidal voltage is applied to four electrodes at its quadrants, the cylinder will vibrate with a nearly pure internal torsional motion. Crystalline quartz is usually selected as the piezoelectric material because of its superior physical properties. When the crystal immersed in a sample fluid, a shear wave will be produced in the sample fluid and will be rapidly attenuated as it moves normal to the surface of the crystal. The viscous drag, exerted by the fluid on the surface of the crystal, changes the crystal's resonant frequency, its conductance at resonance, and its bandwidth at resonance from their value in vacuum. The equation most frequently employed in recent work,

$$\eta = \pi f_e \left( \frac{M}{S} \right)^2 \left[ \frac{\Delta f_e}{f_e} - \frac{\Delta f_o}{f_o} \right]^2 \tag{5}$$

related  $\Delta f_e$ , the crystal's bandwidth at resonance to the viscosity-density product of the sample fluid: viscosity,  $\eta$ , and density,  $\rho$ . Here,  $M$ ,  $S$  and  $f_e$  are the mass, surface area and resonant frequency of the crystal, respectively, while the subscript zero is corresponding to the equivalent quantities in a vacuum area. Diller (1965) has measured viscosity of parahydrogen with Quartz crystal oscillation method for pressures and temperatures up to 30 MPa and up to 100 K, respectively. Schematic diagram of Diller's apparatus is shown in Figure 3. This viscometer gives small repeatable bandwidths in vacuum. A typical crystal has a length of 5 cm, a diameter of 0.3 cm and a 39.5 kHz resonant frequency, which yields a fluid boundary layer of about  $10^{-4}$  cm, whereas the gap containing the fluid is about  $5 \times 10^{-2}$  cm.

**Oscillating body method**

Oscillating-body viscometers consist of an axially symmetric body suspended from a torsional wire so that the body performs oscillations in the fluid about its axis of symmetry. The oscillator can be a disk, a cylinder, a plate, a sphere or a cup. The sample usually surrounds the oscillator but in the case of the sphere or the cup, it can be inside them. The oscillating disk (OD) method was successfully developed by Kestin *et al.* (1959), this method was widely used to measure gas piece. The coefficient viscosity is determined from the period and damping of oscillation and the characteristics of the oscillation system. The method has the advantages of compactness, high precision and accurate in the working equation. Figure 4 shows the Navizadeh viscometer apparatus developed with OD method (Navizadeh and Mayinger, 1999). The OD of  $R = 29.55$  mm in radius and  $d = 1.5$  mm in thickness was suspended by means of platinum-iridium wire of diameter,  $D = 0.06$  mm. A spacer ring made of optically grounded quartz glass ensured the distance between the fixed plates. The clearances between oscillating disk and fixed plates were 1.01 mm. The oscillating system was accommodated in a suspension holder by means of supports of different plates materials and allowed an axial displacement of only  $0.00045 \text{ mm}^\circ\text{C}^{-1}$ .

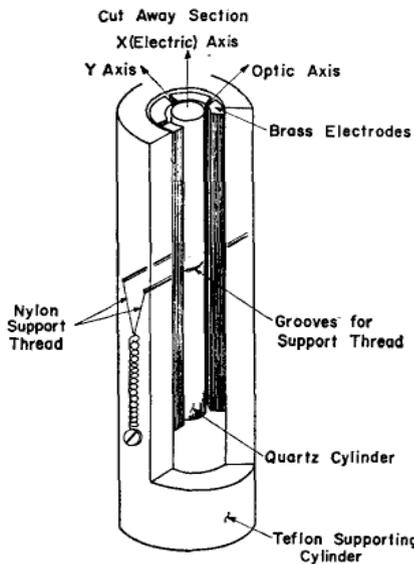


Figure 3. The quartz crystal oscillator developed by Diller (1965)

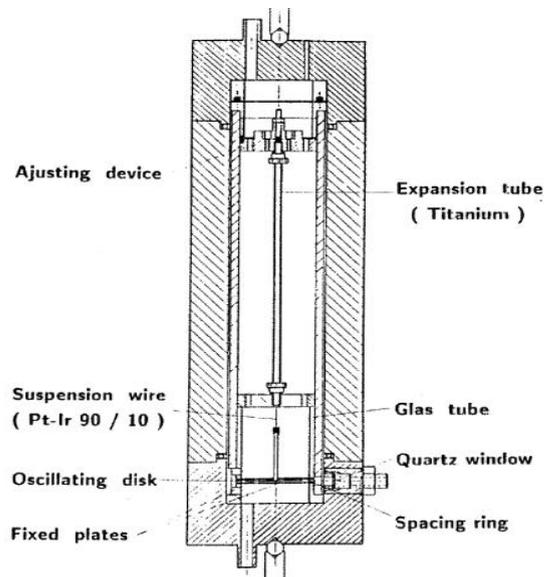


Figure 4. Oscillating disk viscometer used by Nabizadeh and Mayinger (1989)

To determine the coefficient of viscosity, it was necessary to calibrate the viscometer with a reference viscosity in order to obtain the edge effects,  $\delta$ , as a function of boundary layer thickness. It defines as

$$\delta = \sqrt{\eta T_o / 2\pi\rho} \tag{6}$$

where  $\eta$  represents the dynamic viscosity,  $T_o$  is the periods of oscillating in vacuum and  $\rho$  is the density of the sample. According to Kestin (1959), the following working formula for calibrating and evaluating of the present measurement should use the edge-correction factor,  $C$ , as expressed above

$$C = \frac{2(T_o / T) [\Delta(T_o / T) - \Delta_o]}{(\pi\rho R^4 \delta / I) \{H_1 K_2 + H_2 K_1 + (2d / R) [H_1 + 3\delta T_o / 2RT]\}} \tag{7}$$

where  $I$  is the moment of inertia of the suspension system.  $\Delta$  and  $\Delta_0$  are logarithmic decrements of damping in media and in vacuum, respectively.  $T$  is the periods of oscillating in media.  $R$  is radius of the oscillating disk,  $d$  is thickness of the oscillating disk,  $H$  and  $K$  consist of an equation, which it can be seen in detail at Nabizadeh' papers (1989).  $\Delta_0$  and  $T_0$  both depend on temperature and were obtained at temperatures ranging from (303 to 503) K in the viscometer itself. Nabizadeh (1999) obtained the edge-correction factor,  $C$ , by using nitrogen at temperatures ranging from (303 to 473) K and at pressure up to 8 MPa.

**Vibrating method**

The straight vibrating-wire (SVW) viscometer is a particularly simple form of vibrating viscometer, which has successfully been employed to measure over a wide range conditions. The theory of a vibrating wire viscometer can be used to determine the viscosity of a fluid from the frequency and the logarithmic decrement of a damped harmonic oscillation (Retsina *et al.*, 1987). However, the validity of the theory is restricted by conditions of the fluid mechanics, which have to be fulfilled. The fluid has to be regarded as an incompressible continuum. Restina (1989) assumed that the maximum wire displacement related to the wire radius,  $r$  and the Reynolds number are very small, so that the convection term in the Navier-Stokes equations can be omitted. Provided that  $r$  is much smaller than the wire length  $L$ , there is no significant flow along the wire axis and the stream problem becomes planar. The working equations needed to calculate the viscosity in an iterative way are given as

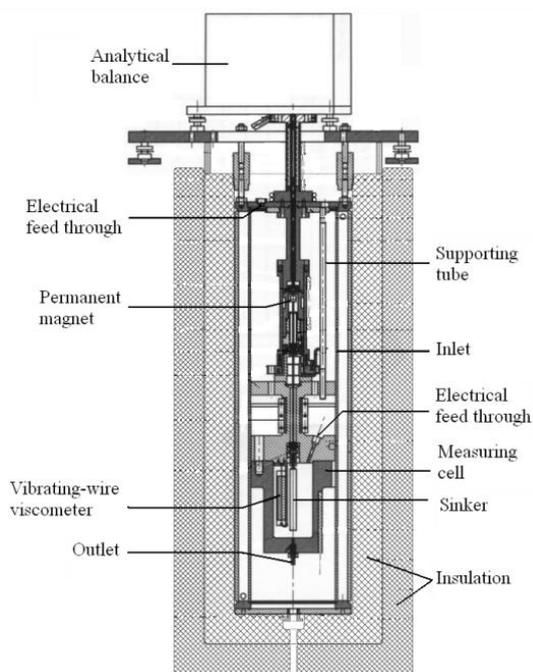
$$\Omega = \frac{\rho \omega r^2}{\eta} \tag{8}$$

$$A = (i - \Delta) \left[ 1 + \frac{2K_1(H)}{hK_0(H)} \right], h = \sqrt{(i - \Delta)K_2} \tag{9}$$

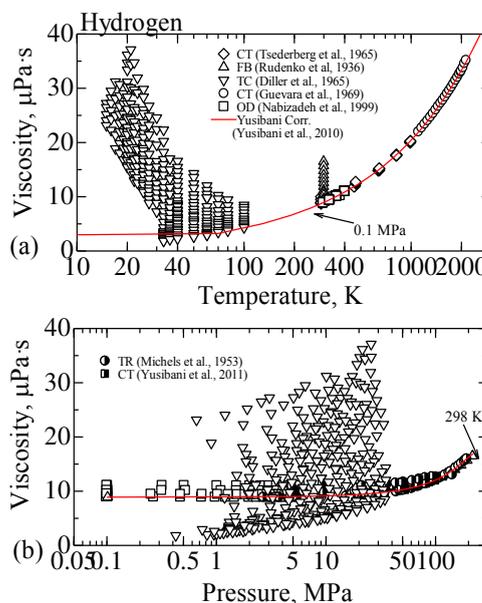
$$k = -1 + 2 \operatorname{Im}(A), k' = \operatorname{Re}(A) + \Delta \operatorname{Im}(A) \tag{10}$$

$$\Delta = \frac{\left(\frac{\rho}{\rho_w}\right)^{k'+\Delta_0}}{\left[1 + \left(\frac{\rho}{\rho_w}\right)^k\right]} \tag{11}$$

where  $\omega$  is the motion frequency,  $\Delta$  is the logarithmic decrement of a damped harmonic oscillation,  $K_0$  and  $K_1$  are modified Bessel functions, respectively,  $\Delta_0$  is the logarithmic decrement in a vacuum condition. The calculation procedure assumes that the density of the sample,  $\rho$ , the density of wire,  $\rho_w$ , the wire radius and the parameter of the oscillation in vacuum are known. Trappenier *et al.* (1980) has measured argon viscosity for temperature up to 323 K and pressure up to 779 MPa with this method. Van der Gulik *et al.* (1988) used this method to measure methane viscosity up to 1000 MPa at 298 K. Wilhelm *et al.* (1998) also applied this method to measure dilute and dense gas. Schematic diagram of Seibt's apparatus is shown in Figure 5. This apparatus can simultaneously measures viscosity and density properties in accurate way.



**Figure 5.** Diagram of compact densimeter-viscometer used by Seibt *et al.* (2009) with straight vibrating wire method



**Figure 6.** Hydrogen viscosity measurements with various methods versus (a) temperature and (b) pressure (CT=capillary tube, FB=falling body, TC=torsional crystal, OD=oscillating disk)

The measuring system consists of a vibrating-wire viscometer and a single-sinker densimeter. This is very attractive method for high pressure region measurements. A major challenge of this method is to apply at high temperature. One short coming with straight wires is that careful attention must be given in the construction, clamping and/or

orientation of the magnetic field to avoid exciting oscillations in two different perpendicular planes having very slightly different resonant frequencies (Trappeniers *et al.*, 1980). One compact, robust and successful strategy to overcome this problem is to use a wire bent into a curve, or part of a circle. The curved wire also has the merit that there is no need to impose a tension on the wire since the elasticity of the wire material can provide the restoring force (Morishita *et al.*, 1989).

## A Comparison of Method and Recommendations

Hydrogen is the most difficult of gas piece to be measured, recently. The coefficient of viscosity-range is small and also the density is the smallest among to the atomic gases piece. Comparison of hydrogen measurement with various methods has been done. High temperature measurement region is done with a capillary tube method with more than 2000 K (Figure 6a), and the high pressure region with falling body method of 200 MPa at atmospheric pressure (Figure 6b). The line is a hydrogen correlation developed by Yusibani *et al.* (2010). The discrepancy of the present correlation is as much as (2 to 4) % with published data selected from the high-temperature and high-pressure region. Temperature and pressure ranges of the present correlation are (40 to 2130) K for dilute gas, and (100 to 990) K for (0.1 to 220) MPa, respectively. The measurement both in the high temperature and high pressure region was done with capillary tube method.

The simplest mechanical construction with a high reliability and a simple equation is the advantages of a capillary tube method. Oscillating disk, torsional crystal and vibrating wire method is value of compactness and required of small amount of sample. For extreme pressure and cryogenic measurement they are the best. The measurement at extreme temperature region is still challenging.

## Conclusions

Many researchers have relied on the capillary tube method to measure gas viscosity at both in high pressure and high temperature. This method has the advantages of simplicity of mechanical construction and operation. However, to have an excellent accuracy with this method both in high pressure and high temperature is by no means an easy task. Oscillating disk method is other alternative to measure gas viscosity. However, literature survey reveals that the feasibility of the method at high pressure is still challenging. Vibrating wire method seems to be a promising method to measure gas viscosity under high pressure region. However, a major disadvantage of this method is the difficulties to apply the instrument at high temperature since this method required magnetic field to measure viscosity.

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