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FLOW DEVICES WITH CORROSIVE GASES**

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AN INSTRUMENT FOR GRAVIMETRIC CALIBRATION OF FLOW DEVICES WITH CORROSIVE GASES

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ABSTRACT

An instrument was developed for the direct mass flow calibration of gas flowmeters that does not require measurement of temperature, pressure, or specific volume. This instrument measures the weight of gas collected in a container and makes measuring those thermodynamic variables unnecessary. The need to measure the weight of the gas container is eliminated by submerging it in a liquid (presently water) and balancing its weight with the force of buoyancy. The accuracy of this Gravimetric Calibrator is unaffected by the pressure and temperature of the gas. The Calibrator can also measure reactive, corrosive, and non-ideal gases. The container remains connected to the process by a torsion capillary, and a load cell measures the changing gas weight continuously throughout the measuring process. A prototype was designed for gas flows ranging from 1 sccm of hydrogen to 10,000 sccm of tungsten hexafluoride, constructed, tested, and used to calibrate flow devices. Experience with the prototype and results are presented, and plans for further developments are discussed.

INTRODUCTION

Many industrial applications require accurate flow measurements of various process gases and vapors. Usually the quantity of interest is the mass flow rate or the equivalent volume flow at some reference condition (i.e. standard temperature and pressure). Most gas flowmeters are influenced by fluid density and other properties such that they cannot directly measure the mass flow rate. The process pressure and temperature must also be measured so that an appropriate "density" correction can be made on the flowmeter signal. There are two types of gas flowmeters, which can measure mass flow directly without the need for density corrections. These are thermal flowmeters and Coriolis flowmeters. Coriolis meters, which are based on rotational dynamics, have been available for liquids for many years and are just becoming available for gas flow, generally in the higher ranges. Thermal flowmeters, which are based on convective heat transfer effects, are very popular because they can provide accurate and economical mass flow measurements over a wide range of flow rates and process conditions .

A significant advantage of thermal flow meters is that they can be calibrated at ambient conditions on a specific gas and then be used at process conditions to make accurate mass flow

measurements without the necessity for monitoring pressure and temperature in order to make density corrections. If the process gas is highly reactive or toxic, it may be very difficult or impossible to perform a calibration, even at ambient conditions. In these cases it is a common practice for flowmeter manufacturers to calibrate the meter on a substitute (or surrogate) gas, which is safer to handle and matches the thermal characteristics of the process gas as closely as possible. Unfortunately, because of the complexity of the thermal processes within the flow sensor, there is no accurate and straightforward method for predicting the performance on one kind of gas based on the calibration on another gas. The use of a surrogate calibration gas instead of the actual process gas can result in large errors. Because of the widespread use of thermal meters, especially in the semiconductor industry, there is strong motivation for mass flow calibrators, which can accommodate a variety of process gases.

Traditional gas flow calibrators are not designed to measure mass flow directly or to be operated with corrosive or toxic vapors. Some of the traditional methods and apparatus for calibrating gas flowmeters are discussed briefly below.

Volumetric Gas Flow Calibrators

Volumetric calibrators determine the rate of mass flow indirectly from measurements of temperature, pressure, and volume using the equation of state for ideal gases. One of the most common types is illustrated in figure-1:

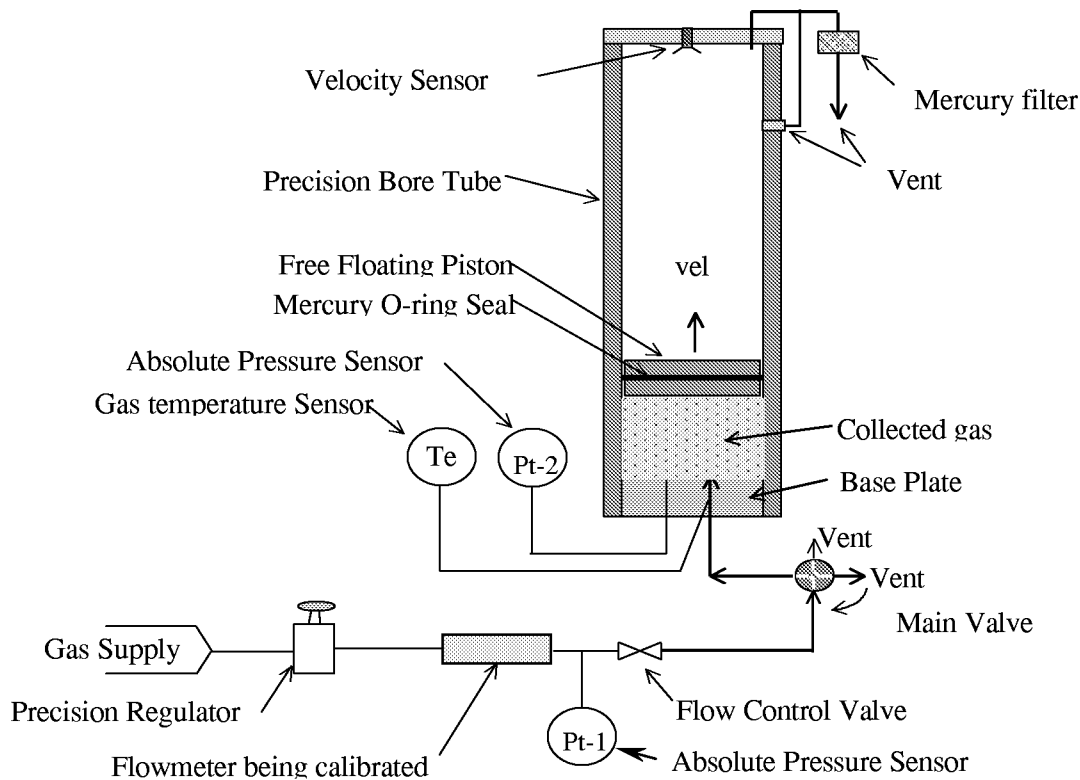


Figure-1: Constant Pressure Volume Flow Calibrator

In figure-1 the gas flows through the meter being calibrated and then is collected in a precision bore tube under a rising piston. By measuring the rate of rise of the piston, the volume flow rate of the gas can be accurately determined. By measuring the pressure and temperature beneath the piston the density of the gas, and thus the mass flow rate, can be calculated. A variation of this system, instead of a moving piston, is to collect the gas in a vessel of constant volume, in which the internal volume is accurately known. By measuring the rate of pressure change the mass flow rate of the gas can be calculated with an appropriate equation of state.

Volumetric calibrators are capable of accuracies approaching a few parts per thousand if the collection pressure and temperature are accurately measured, and if the equation of state is sufficiently accurate. If the gas or vapor is non-ideal, or reacts in any way with the walls of the collection vessel, then large errors in the mass flow determination can occur. For this reason volumetric systems are not suited for many process gases.

Weight -Time Gas Flow Calibrators

The most direct mass flow calibration method would be to directly measure the mass of the gas as it is collected in a vessel. A system which weighs the collected gas offers an advantage over volumetric systems in that the mass flow rate is measured without having to perform density corrections. Because density corrections produce additional uncertainties, direct weighing systems are potentially more accurate than volumetric systems. Unfortunately, due to the relatively low density of most gases, very large quantities must be collected for an accurate weight measurement. Although there are electronic balances which can detect microgram or nanogram quantities, they are limited in capacity to a few grams or milligrams. A collection vessel for even small quantities of gas will usually weigh several kilograms. Therefore, a few milligrams (or micrograms) of gas must be weighed in addition to the large tare weight of the vessel. This means that very large quantities of gas must be collected for accurate weighing, which requires very high flow rates or very long collection times. A method for performing gas flow calibration by direct weighing is illustrated in figure-2.

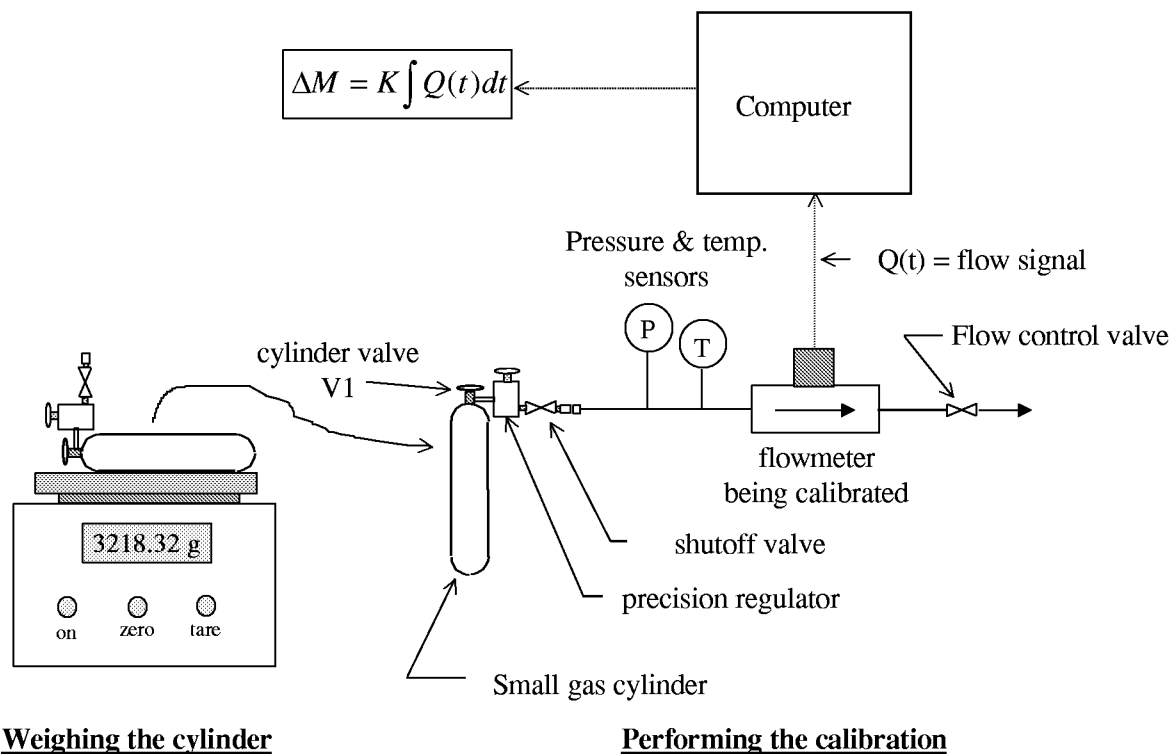


Figure-2: Gas Flow Calibration by Direct Weighing

In figure-2 the small gas cylinder is pressurized with the desired calibration gas and a good pressure regulator and shutoff valve are attached as shown. The flowmeter to be calibrated is arranged as shown with a flow control valve downstream (or upstream) of the meter. The gas cylinder is first connected to the flowmeter piping manifold and the regulator and flow control valve are adjusted to give the desired pressure and flow rate for calibration. The desired flow is indicated by the flowmeter signal. The shutoff valve is then closed and the cylinder – regulator assembly is disconnected and placed on the electronic balance for an initial weighing.

After weighing, the cylinder is reconnected to the manifold. The shutoff valve is opened to start the calibration flow and the computer integrates the flowmeter signal for the duration of the calibration run. The run must continue until sufficient gas has left the cylinder to obtain an accurate weighing. For example, if the accuracy (or resolution) of the balance is 50 milligrams, you will have to discharge 50 grams of gas to obtain a 0.1% weighing uncertainty. If the gas is nitrogen for which the standard density is approximately 1.15 grams per liter, and if the flowrate is 0.5 standard liters per minute, then a run time of approximately 90 minutes is required. At the end of the run the shutoff valve is closed and the cylinder – regulator assembly is disconnected and re-weighed to obtain the total mass of the gas discharged during the run. The initial and final mass values along with the integrated flow signal can be used to calculate the calibration factor (K) for the flowmeter as shown in figure-2 . With careful operation this procedure is capable of determining the meter calibration factor with an uncertainty of less than one part per thousand.

There are several disadvantages to this direct weighing method. Because of the large quantity of gas required for accurate measurement, the collection times can be several hours or more, especially at low flow rates. It can take many days to calibrate a flowmeter if several different flow rates are needed. The procedure is further prolonged because the cylinder has to be

disconnected from the system for each weighing. If the process gas is hazardous, the pipes communicating with the cylinder have to be purged before the connection to the cylinder is opened

Motivated by a need for more accurate and reproducible mass flow measurements of a variety of corrosive and toxic process gases in the semiconductor The Gravimetric Calibrator discussed in the following section was developed to eliminate the most restrictive limitations of the calibration methods mentioned above.

DESCRIPTION OF THE GRAVIMETRIC CALIBRATOR

The Gravimetric Calibrator measures the mass of a quantity of gas collected in or discharged from a receptacle, or it determines the mass flow rate. It achieves this by measuring the weight of the gas. Figure 3 shows a schematic diagram of the instrument.

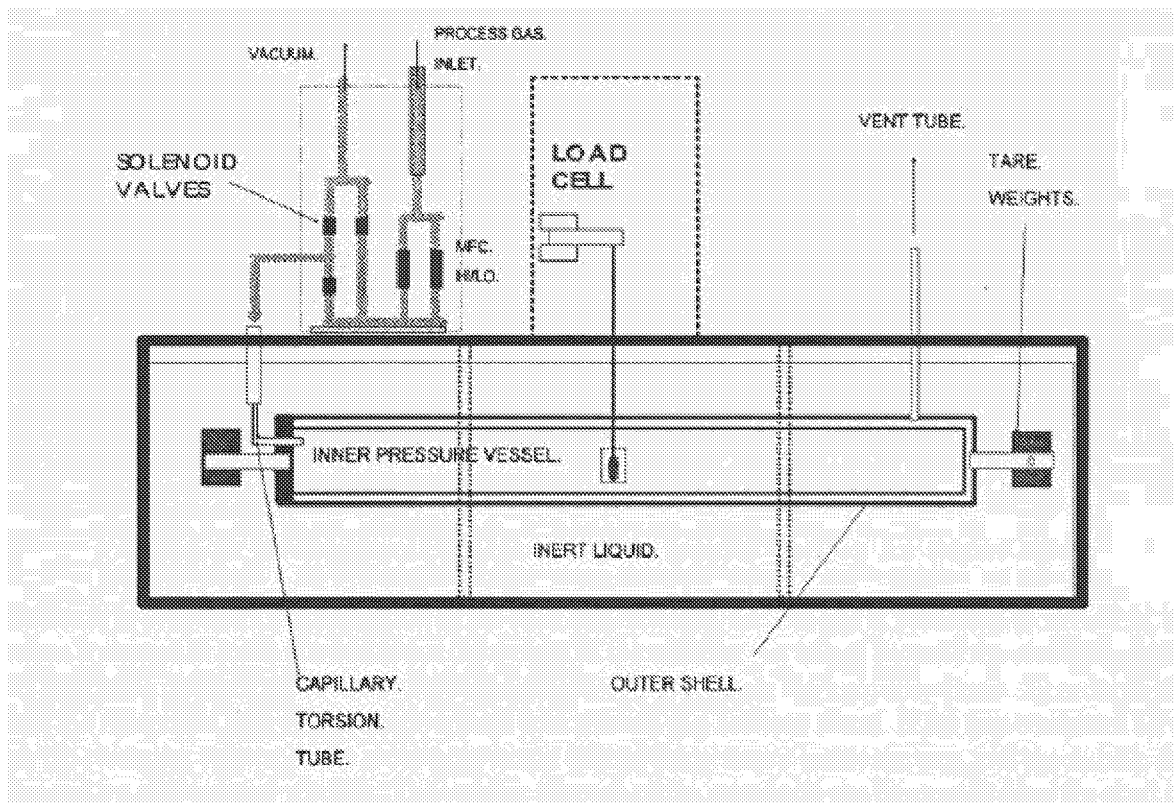


Figure-3: Configuration of Gravimetric Calibrator

The receptacle is a long cylindrical container submerged in water with its long axis horizontal. The internal volume is approximately 20 liters and the weight is approximately 50 Kg. Its weight is adjusted so that it takes a force of a few grams to prevent it from sinking when it is evacuated. The receptacle is supported at two points along a horizontal axis that is perpendicular to the long axis, and it goes through the centroid of the internal cavity of the receptacle. When

there is any amount of gas inside the receptacle, the center of mass of this body of gas coincides with the centroid. Thus, this configuration insures that any gas inside the receptacle is supported through its center of mass. The significance of this will be explained later. The receptacle is connected to the gas flow system through a capillary tube. This capillary tube is horizontal and parallel to the transversal axis of the receptacle. In this arrangement, the capillary is subjected to torsion when the receptacle centroid moves up or down.

The receptacle is suspended from a load cell. If the receptacle is evacuated, the indication of the load cell recorded, and the gas is permitted to flow in, the load cell indicates a change in load. But this is not the weight of the entering gas alone, because the changing pressure inside the receptacle changes the receptacle's dimensions and that changes the buoyancy force. Although these changes are very small, they are significant in comparison to the weight of the gas being collected. In order to eliminate this effect, the receptacle was placed inside a shell with a small gap between them. The shell is vented to the atmosphere through a capillary tube so that the gap contains air at atmospheric pressure. This effectively de-couples the receptacle volume changes from the shell, however there is still a possible source of error. With this arrangement, changes in barometric pressure can change the amount of air in the gap. Since the air in the gap is also weighed by the load cell, changes during a calibration run would cause a "zero shift" and could result in errors in the mass flow calculation. It turns out however, that the barometric pressure changes that normally occur during a calibration run produce an insignificant error in the weighing process.

OPERATION & CONTROL OF THE GRAVIMETRIC CALIBRATOR

The gravimetric calibrator that is currently in use has a receptacle (vessel) with a 20 liter capacity. It is designed to operate over a range from 1 sccm of hydrogen (Molecular Weight = 2) to 10000 sccm of tungsten hexafluoride (Molecular Weight = 298). The goal was to provide a mass flow rate accuracy of 0.1% or better over this range. The calibrator was thoroughly tested on nitrogen to determine its performance capability, and has subsequently been used with a variety of process gases common to the semiconductor industry. Once a few startup problems were solved, the system was found to be capable of the 0.1% accuracy goal for nitrogen flows of 100 sccm and above. Below 100 sccm the error (as a percent of flow) increases from 0.1% up to about 1% at 10 sccm. Figure 4 illustrates the valving and control arrangement for the calibrator. The basic operation and control of the system is described below, along with some of the startup problems and sources of error.

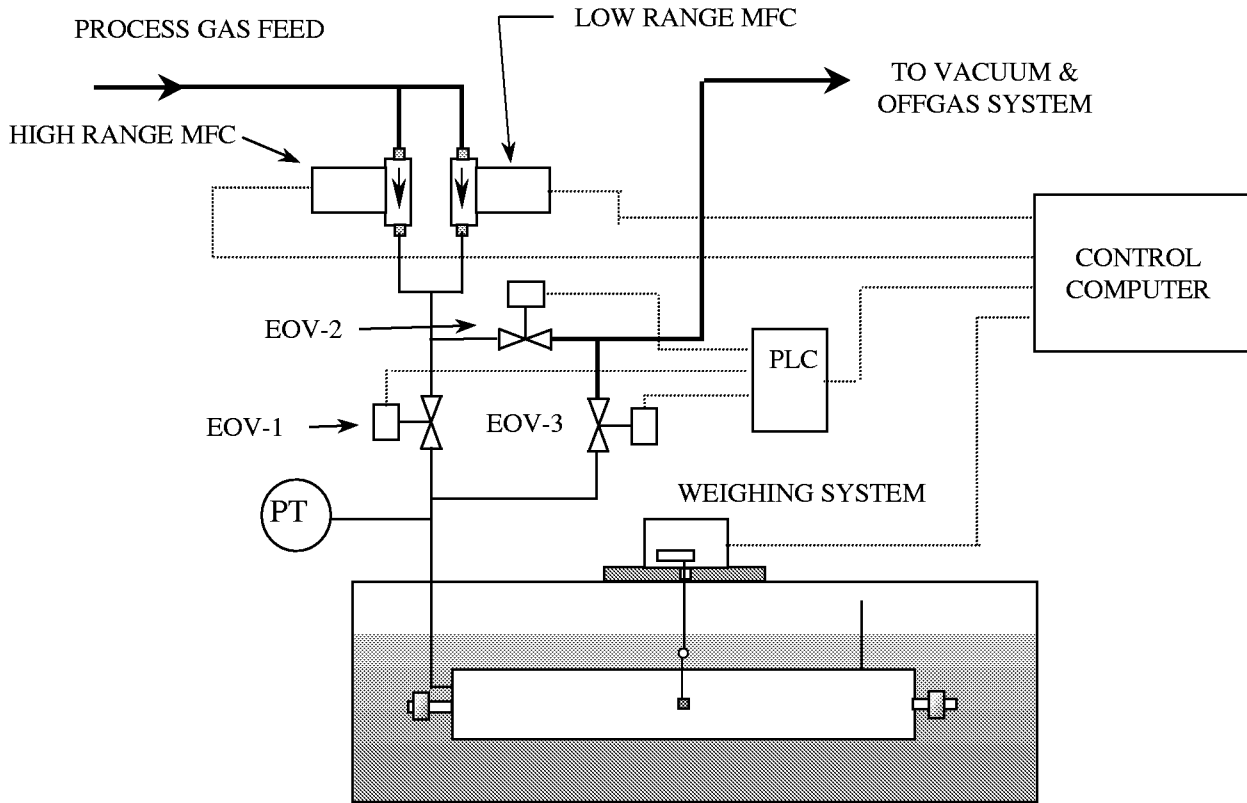


Figure-4: Gravimetric Calibrator Control System

Operation of the Gravimetric Calibrator

The weighing system uses a 50 gram strain-gage load cell, and is calibrated to weigh about 20 grams of gas. The weighing system is calibrated in place by placing small weights on a platform attached to the vent tube assembly. An analog IO board in the control computer reads the load cell signal. The process gas is supplied through two thermal mass flow controllers (MFC) arranged in parallel as shown. The high range MFC is 10000 sccm and the low range MFC is 1000 sccm. The inlet valves EOV-1, EOV-2, and EOV-3 are small fast acting 2-way solenoid valves and are operated by a programmable logic controller (PLC) which interfaces to the control computer. A calibration is performed as follows:

The device being calibrated is installed in the process gas feed line upstream of the MFCs. Before process gas flow is started, EOV-1 and EOV-2 are closed and the collection vessel is evacuated through EOV-3. When the desired starting pressure is reached, EOV-3 is closed and the collection vessel pressure and weight are allowed to stabilize. To start the calibration, EOV-2 is opened and one of the MFCs is set to the desired flow rate. Process gas flows through the device being calibrated, the MFC, and EOV-2 to the vacuum / offgas treatment system. To begin a calibration run, the operator presses a "start key" on the control computer, and the PLC closes EOV-2 and opens EOV-1. EOV-3 remains closed. The gas begins to flow into the collection vessel. The computer records the starting time and weight and the operator allows the flow to continue for the desired collection period. The collection periods normally range from about 30

seconds to about 3 minutes, depending on the flow rate. At the end of the collection period the operator presses the “stop key” and the PLC closes EO V-1 and opens EO V-2. The computer records the final time and weight and computes the average mass flow rate. During the collection interval the weight and time are logged about 5 times per second and the slope (“moving” least squares) of the weight verses time line is computed continuously and displayed on the screen. The slope is a continuous indication of the mass flow and will agree with the end point calculation if the flow rate was constant over the collection interval. Closing EO V-2 and opening EO V-1 starts the next calibration run. The vessel is not evacuated between runs. It only needs to be evacuated when the total mass of gas reaches the measurement limit of the weighing system. With nitrogen, this occurs at a pressure of about 1 atmosphere in the vessel.

Startup Problems and Sources of Error

There were several sources of error that had to be eliminated in order to obtain the desired system performance. Some were anticipated in the design and some were discovered during the initial testing. These are reviewed briefly in this section.

Use of an Electronic Microbalance Weighing System. The original plan was to use an electronic micro-balance (such as the Mettler AT-201) as the weighing device. The balance was to sit in an enclosure above the water tank and connect to the vessel by means of a thin rod connected to the bottom of the weighing pan. When the balance was turned on it experienced severe vibrations which could not be damped out. After a few days of trying to eliminate the vibrations, it was observed that the vibrations were so large that the motion of the vessel was visible to the eye, and that the vibrations stopped whenever the balance was turned off. It turned out that the weighing mechanism of the balance uses a position feedback servo loop, which was unstable with the extra mass of the vessel attached, even though the weight was balanced by buoyancy. Rather than attempt to modify the electronic balance, it was decided to replace it with a low range (50g) load cell.

Zero Drift Corrections. At the beginning of a calibration run, with the vessel isolated, a zero flow calibration run is performed. Ideally the measured flow should be very close to zero. This was not the case during initial testing. Very large “zero” flow rates (100 to 500 sccm) were observed. Of course, if this “zero” flow remains constant during a cal run it can be subtracted from the calibration flow. With such a large zero correction however, the error in measuring the zero becomes significant. It was soon discovered that the “zero flow” seemed to track the normal changes in room temperature over the course of a 24 hour period. Even though the temperature changes in the water were very small, the change in water density was enough to produce significant changes in the buoyant force and therefore the observed “zero” flow rate. A change in water temperature of 1 degC results in a 5.5 gram change in the buoyant force. This problem was solved by enclosing the water tank in a plywood box and controlling the air temperature in the box at 20 degC. An alternative solution that was considered was to keep the water at 4 degC where the density change with temperature is zero. Since the first solution was adequate, the second solution has not yet been tried.

Forces Exerted on the Vessel by the Torsion Capillary. When the receptacle is in some general position, the capillary is strained in an unknown way and exerts a force on the receptacle and thus affects the load cell indication. If the receptacle does not move, this force is constant and has no effect on the measurement. The changing gas weight, however, changes the deformation of the load cell and the suspension mechanism resulting in a vertical displacement of the receptacle

and consequently also in a change in the capillary force. This force change is proportional to the change in the gas weight. The error can be calculated or determined by calibration to make corrections, but, since the capillary is designed so that the force in question is four to five orders of magnitude smaller than the weight of the gas, the error is negligible.

Effect of the Center of Mass of the Gas. The location of the receptacle suspension has to be measured carefully. If the line of action of the load cell reaction goes through the mass center of the gas, the entire gas weight acts on the load cell. If the mass center is between the capillary and the suspension, the weight of the gas is distributed between the capillary and the load cell, and the load cell indication is less than the weight. If the mass center is farther away from the capillary than the suspension, the indication is more than the gas weight. This deviation is again proportional to the gas weight; it can be calculated from the dimensions of the receptacle or determined by calibration, and a calibration factor can be calculated if the deviations are too large to be ignored. This is not a flaw in the instrument, and special applications may make it preferable to locate the suspension deliberately off the mass center.

The location of the center of mass of the fluid being weighed is not a problem as long as it remains constant. This will always be the case as long as the fluid remains a gas and any reaction with the vessel walls is fairly uniform over the internal surface. However, if the fluid is a condensable vapor, and condensation forms at various places in the vessel, the center of mass of the fluid may shift and produce a significant weighing error. If the location of the center of mass is unknown, then the effect on the weighing system is statically indeterminate and cannot be corrected by analytical means. A solution to this problem has been developed. The solution involves the use of three load cells (instead of one) and an algorithm, which computes the location of the mass center and the weight of the fluid. The three load cell system has not yet been implemented.