Calibrating flowmeters for use in measuring contamination in high-purity process liquids

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A correlation procedure helps predict calibration curves for flowmeters used in assessing particle concentrations in UHP chemicals.

The cleaning and etching processes used to manufacture semiconductor microcircuits require numerous steps employing a variety of hazardous liquid process chemicals. Many of these steps are adversely affected by the high levels of particulate contamination typically found in liquid chemicals. Particle contamination in these chemicals is usually measured using optical particle counters that report particle concentration as the number of particles per unit volume (traditionally, particles per milliliter or per liter). The accuracy of the particle concentration depends on accurately measuring the flow of the liquid chemical through the particle counter. Float-type variable-area flowmeters are often used to measure the flow rate. Because of the corrosive nature of semiconductor process liquids, the flowmeter body and float must be made of Teflon PFA or another inert material.

Ideally, since a flowmeter's calibration depends on the physical properties of the liquid flowing through it, each meter should be calibrated in each process liquid. However, flowmeter manufacturers typically supply a calibration for water only, so the user must recalibrate the flowmeter for each specific liquid chemical. Such recalibrations are hazardous, time-consuming, and expensive. The purpose of the study reported herein was to derive a correlation that allows a calibration curve to be developed for a particular flowmeter if the density and viscosity of the liquid are known. This study does not attempt to mathematically solve the complex boundary-layer problem in variable-area flowmeters using fundamental fluid mechanics principles. Due to the wide variety of flowmeter and float designs, any attempt to derive a complete solution would require too many assumptions and approximations to be of practical use.

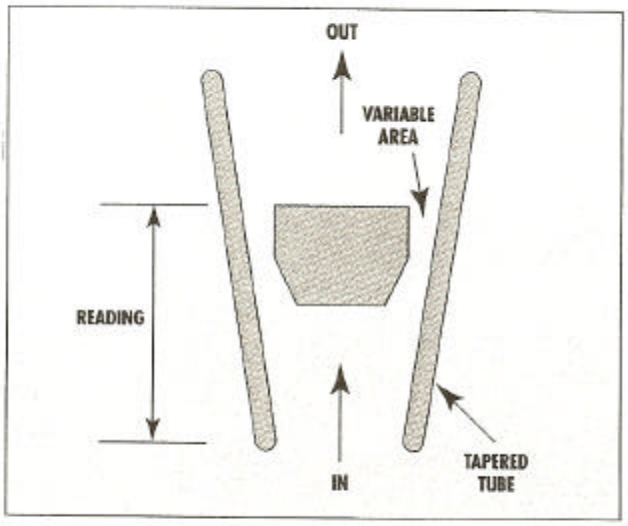

Figure 1: Simplified schematic of a float-type variable-area flowmeter.

Theoretical Background

Float-type variable-area flowmeters consist of a vertical, linearly tapered tube, with the smaller-diameter end at the bottom, through which the fluid flows upward (Figure 1). The float is slightly narrower than the lower end of the tube and rests at the bottom of the tube when there is no flow. The float can move vertically, exposing a variable area between itself and the walls of the tapered tube. In response to the liquid flow rate, a position is reached where the weight of the float, less its buoyancy in the liquid, is balanced by the inertial and drag forces of the fluid acting on the float. Flow rate is indicated by the position of the float's reading edge against a scale on the tube.

The term *float-type* describes those variable geometry devices in which the external force is provided wholly by gravity. In order to operate correctly, these devices must remain vertical. Dimensional analysis has been used to derive an equation for the flow rate (q) for variable-area flowmeters[1] (Equation 1):

$$q = CD_f\sqrt{\frac{F}{\rho}}$$

In Equation 1, D_f (largest float diameter), F (restoring force tending to zero the float), and ρ (fluid density) are constant for any one flowmeter. For a given liquid-flowmeter pair, C (simplified flow parameter) varies primarily with the float elevation in the tube and F (fluid viscosity) is a constant. When measuring the flow of incompressible liquids, C is a function of N (viscosity parameter), where N is defined by Equation 2:

$$N = \frac{\sqrt{F\rho}}{\mu}$$

The complete working data for a particular variable-area flowmeter for incompressible liquid flow consist of a family of curves of C versus N, each curve representing a particular value of scale reading as a function of N.

The equations given thus far are applicable to any variable-area flowmeter, regardless of the nature of the external restoring force attempting to move the float toward zero. In the case of float-type variable-area meters, which employ gravity for the restoring force, the influence of fluid density in buoying the float must be considered. The value for F is therefore determined by Equation 3:

$$F = gM_f\frac{\rho_f-\rho}{\rho_f}$$

where g is acceleration of gravity, M_f the mass of float, and ρ_f the density of float.

The working equations for C and N, applicable to those variable-area flowmeters employing float shapes within prescribed limits, are subsequently defined as Equations 4 and 5:

$$C = \frac{q}{f_g f_b D_f\sqrt{gM_f\frac{\rho_f-\rho}{\rho_f\rho}}}$$

$$N = \frac{\sqrt{gM_f(\rho_f-\rho)\left(\frac{\rho}{\rho_f}\right)}}{\mu}$$

In order to increase the accuracy of predicting flowmeter performance for a liquid of known density and viscosity, we have chosen to establish the flow characteristics for individual flowmeters, rather than for a family of like meters. The evaluation of individual flowmeters eliminates the need to determine the flowmeter dimensions of D_f, f_b (dimension of float body), and f_g (dimension of bore of float guide), and allows a modified form of the flow parameter (C_m) to be defined as follows in Equation 6:

$$C_m = CD_f f_b f_g$$

Thus Equation 4 becomes Equation 7:

$$C_m = \frac{q}{\sqrt{gM_f\frac{\rho_f-\rho}{\rho_f\rho}}}$$

Experimental Methods

Nine flowmeters (Pathfinder, Odyssey series, Futurestar, Edina, MN) were used in this study. Flow rates with ultrapure water (UPW) ranged from 20 ml/min to approximately 25 L/min, with float weights varying from 1 to 140 g and float densities ranging from 2 to 12 g/cm^3. Floats were made from either Teflon or metal encapsulated with Teflon. The flowmeters were mounted vertically in test stands.

The following electronic-grade chemicals, representing a wide range of liquid densities and viscosities, were used:

- High-purity deionized (DI) water.

- 96% sulfuric acid.
- 85% phosphoric acid.
- 70% nitric acid.
- 37% hydrochloric acid.
- 29% ammonium hydroxide.
- Isopropyl alcohol (IPA).
- Acetone.

A 15-L Teflon-lined pressure tank was employed to dispense all chemicals except UPW. For UPW, the flowmeter test stands were connected directly to a UPW supply and flushed to drain. Process chemicals were collected downstream from the flowmeter test stands and poured back into the 15-L tank.

We used a graduated cylinder and stopwatch for evaluating flowmeter performance. A process chemical was loaded into the pressure tank, the lid was closed, and the tank was pressurized with clean, dried, and filtered nitrogen. Each meter on the test stand had an integral needle valve that allowed the float to be set at a chosen mark. The graduated cylinder was placed in the liquid chemical stream and the stopwatch started simultaneously. After sufficient time had elapsed, the graduated cylinder was removed and the watch stopped. Liquid volume, time, and flowmeter mark were all recorded. All even-number flowmeter marks were evaluated in this manner whenever there was enough liquid chemical flow. Each flowmeter mark was measured two or three times and the average value was recorded. The temperature of each chemical was recorded, since we anticipated that meter performance would depend on viscosity, which in turn would depend on temperature. The density of each chemical was also measured. Those chemical and physical properties are listed in Table I.

Observed Flowmeter Performance

Equation 1 predicts a linear relationship between flow rate and equidistant flow marks. As a general rule this proved to be the case, but not for all the liquid chemicals or flow rates. Figure 2 shows typical plots of flow rate versus flow mark for the chemicals tested.

Regression analysis was used to determine the degree of fit and to identify any gross errors in the data collection. The general form for a second-order regression equation is:

$$y = b_0 + b_1x + b_2x^2$$

where y equals the flow rate, x equals the flow mark, and b_0, b_1, and b_2 are the regression coefficients. In a first-order regression the term $b_2x^2 = 0$ and drops out. The regression coefficients can be utilized to predict flow rates at flow marks that were not used during testing. A regression analysis also provides a value for the correlation coefficient. A value of 1 indicates a perfect fit, whereas a value of 0 indicates no relationship between parameters. For the range of flowmeters tested, all but one had a correlation coefficient >0.99, indicating that very good correlations were obtained.

In general, a straight-line, first-order regression plot could be adequately fitted for the chemicals (Figure 2a), except for sulfuric and phosphoric acids. However, the lower the flow rate was, the less linear the plot. In the case of sulfuric and phosphoric acids, a second-order regression curve-fitting routine was needed to fit curves to the data (Figure 2b). For one particular low-flow-rate meter this generalization did not hold true, and second-order regression analysis had to be used to adequately fit the data for all liquid chemicals (Figure 3).

Chemical	Temp. (°C)	Density (g/cm³)	Viscosity (poise)
DI water	25	1.00	0.0095
96% sulfuric acid	25	1.84	0.2060
85% phosphoric acid	25	1.70	0.5290
70% nitric acid	25	1.39	0.0225
37% HCl	22	1.18	0.0210
29% NH_4OH	15	0.90	0.0130
IPA	25	0.80	0.0205
Acetone	18	0.79	0.0035

Table I: Physical properties of the chemicals used to evaluate flowmeter performance.

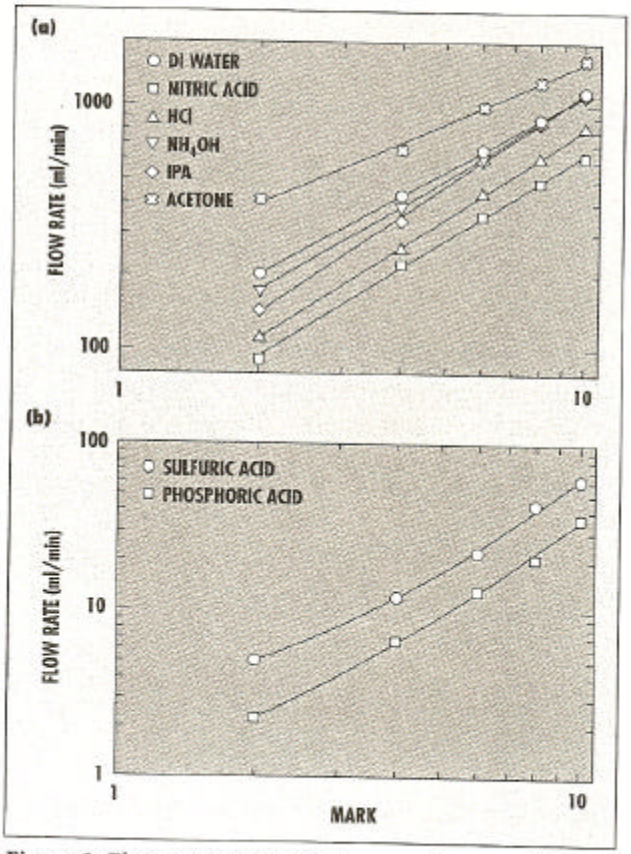

Figure 2: Flowmeter calibration curves in several chemicals with a variety of physical properties.

Predicted Flowmeter Performance

A regression analysis of the raw data showed that the collection method produced consistent and reliable results. These results were then used to predict flowmeter performance for liquid chemicals other than those tested.

The eight chemicals chosen for this study provide a wide range of densities and viscosities. The validity of the suggestion that a family of curves of the modified flow parameter C_m versus the viscosity parameter N (with each curve representing a particular value of scale reading) will provide complete calibration can be tested.

Using Equation 7 to calculate C_m and Equation 5 to calculate N, a series of curves was plotted for each flowmeter at the measured scale mark. Figure 4 depicts an example of a family of such curves for a typical meter. Standard regression analysis proved inadequate to fit curves to the data. Instead a *logistic dose response* equation was chosen. The general form of this equation is:

$$y = a + \frac{b}{\left[1 + \left(\frac{x}{c}\right)^d\right]}$$

where a, b, c, and d are the curve-fitting coefficients.

Substituting for $y = C_m$ and $x = N$ gives the following equations (known as Equation 8):

$$C_m = a + \frac{b}{\left[1 + \left(\frac{N}{c}\right)^d\right]}$$

$$\frac{q}{\sqrt{gM_f \frac{\rho_f - \rho}{\rho_f \rho}}} = a + \frac{b}{1 + \left\{\frac{\sqrt{gM_f(\rho_f - \rho)\left(\frac{\rho}{\rho_f}\right)}}{c\mu}\right\}^d}$$

$$\therefore q = \sqrt{gM_f \frac{\rho_f - \rho}{\rho_f \rho}} \left[a + \frac{b}{1 + \left\{\frac{\sqrt{gM_f(\rho_f - \rho)\left(\frac{\rho}{\rho_f}\right)}}{c\mu}\right\}^d}\right]$$

All flowmeters tested indicated that C_m rises with increasing N. The rate of change is rapid for small values of N and slow for large N values. The curves show the expected properties of asymptotically approaching a constant flow value at higher values of N and a sharp drop-off at lower values of both N and C_m. Depending on flowmeter design, the drop-off can become steeper as the flow-viscosity parameter decreases, as shown in Figures 5 and 6.

For the flowmeters tested, the modified flow parameter rose with increases in the viscosity parameters, changing rapidly at lower viscosity values.

Figures 4–6 indicate that flowmeter performance can be highly viscosity dependent. For example, Figure 4 indicates that the modified flow parameter at Mark 4 varies from 20

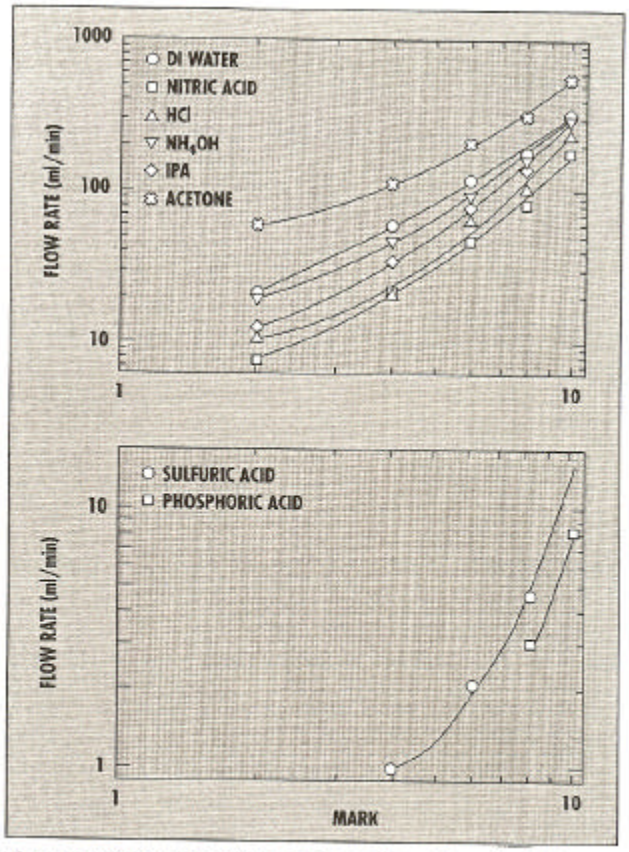

Figure 3: Flowmeter with nonlinear flow characteristics.

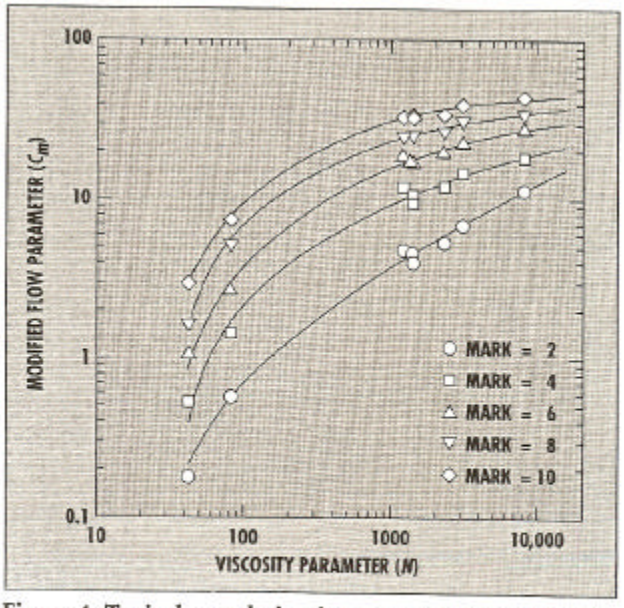

Figure 4: Typical correlation between the viscosity parameter (N) and the modified flow parameter (C_m).

for a viscosity parameter of 10,000, to 0.5 for a viscosity parameter of 40. Hence, if density and viscosity corrections are not taken into account, the flow rate will be off by a factor of 40. Obviously, viscosity corrections are needed to obtain accurate flowmeter calibration curves.

The curves depicted in Figures 4–6 represent the complete calibration, over the range of viscosities tested, referred to previously.[1] The only unknown is the flow rate corresponding to the flow mark. The flow rate corresponding to a particular flow mark on any of the flowmeters tested for a liquid chemical of known density and viscosity can be calculated by substituting the appropriate values into Equation 8.

The flow rate corresponding to a particular flow mark for a liquid chemical of known density and viscosity can be calculated using the logistic dose response equation.

Error Analysis

Figure 7 presents the deviations between the experimental data and the flow rate predicted by the correlations, as a function of the viscosity parameter. The figure was prepared using data from all of the flowmeters tested. The error bars associated with each point represent the 95% confidence limit of the error at a given value of N. The two curves shown in the figure represent mathematical fits of the mean and 95% upper confidence limits.

Figure 7 indicates that the error decreases with an increasing viscosity parameter. This trend is expected since Figures 4–6 indicate that small changes in the viscosity parameter have a large influence on the flow parameter for small values of N, while the effect is much smaller for large values of N. The average error is <10% for viscosity parameters >150, and the 95% upper confidence limit error is <10% for viscosity parameters >900. The error increases rapidly for viscosity parameters <100.

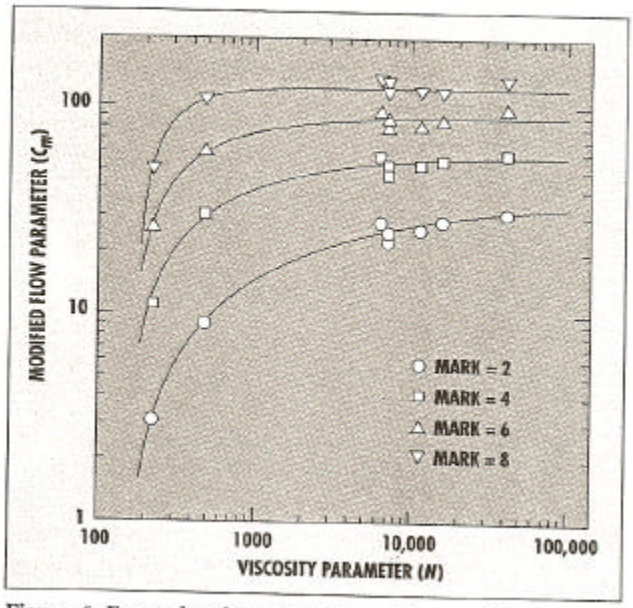

Figure 6: Example of a correlation with a rapid change in C_m at low N values.

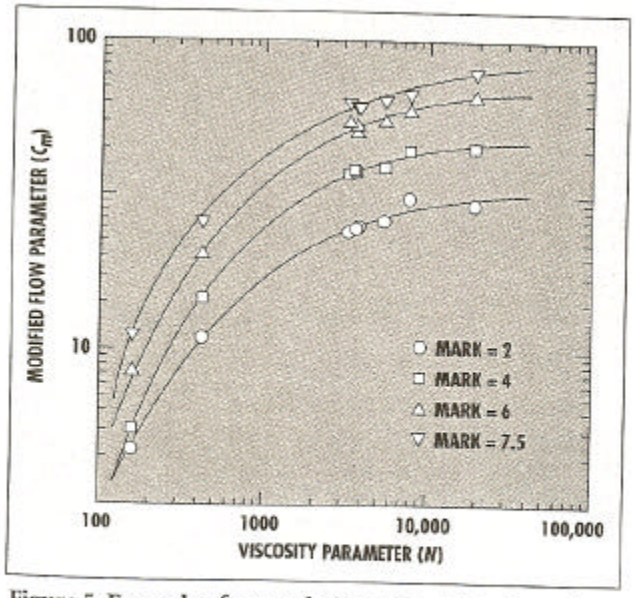

Figure 5: Example of a correlation with a high-dependence of C_m on N.

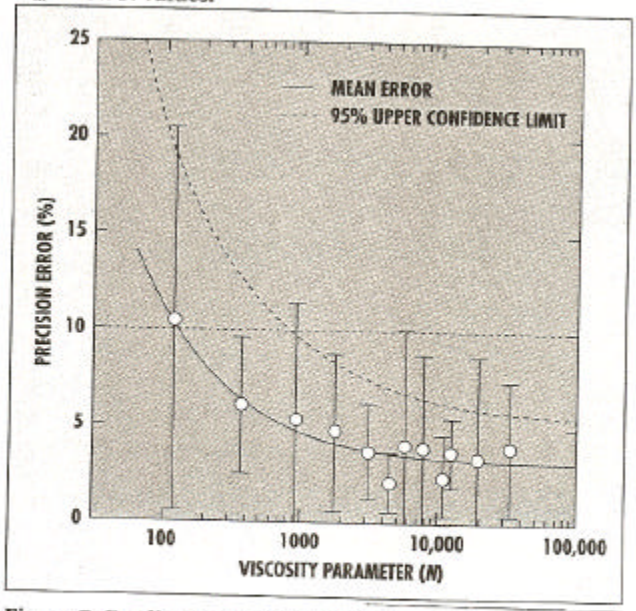

Figure 7: Predictive accuracy of the correlation as a function of N.

Conclusion

A flowmeter's calibration depends on the density and viscosity of the liquid being measured. A large difference in liquid physical properties can result in large differences in flow rate at a given flowmeter mark. For example, the flow rate for chemicals like acetone (low density and low viscosity) can be ≥40 times the rate of phosphoric acid (high density and high viscosity) at the same mark. The flowmeter calibration in different liquids can be predicted using a viscosity parameter, which is a function of the physical properties of the liquid and the flowmeter float. For high-viscosity fluids, the calibration curve was found to be highly dependent on fluid density and viscosity. For low-viscosity fluids, the calibration was highly dependent on the fluid density but less dependent on the fluid viscosity. The correlation procedure used in this article was shown to predict calibration curves for the flowmeters included in this evaluation within 10% (with 95% confidence limits), when the viscosity parameter N was >900. The error increases with a decreasing viscosity parameter.

Acknowledgment

This article is a revised version of a paper presented at the 41st Annual Technical Meeting of the Institute of Environmental Sciences, Anaheim, CA, May 2, 1995. Used with permission.

Reference

1. Head VP, "Coefficients of Float-Type Variable-Area Flow Meters," *American Society of Mechanical Engineers Transactions*, 76(1):854–862, 1954.

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