Properties for Accurate Gas Flow Measurements

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Abstract: Accurate gas properties are needed to take full advantage of the low uncertainties provided by NIST's Gas Flow Calibration Services. If a flowmeter user and NIST use different values for these properties (molecular mass, compressibility, density, viscosity, and critical flow factor), the user's flow measurements will have errors. Calibrations conducted by NIST's Fluid Metrology Group use the NIST-supported database REFPROP version 9 to reduce calibration data. Prior to 2009, NIST's calibration data were reduced using REFPROP 7.0. Flowmeter users who recalibrate their meters at NIST and ignore the January 2009 change of database may erroneously conclude that either their meters or NIST's standards are not stable.

The newer database, REFPROP v. 9, calculates properties for mixtures that include up to 2 % water vapor. By measuring the dew point temperature of air used during our calibrations and by incorporating the REFPROP v. 9 dynamic link library (REFPROP.dll) into our Labview[*] and Excel data acquisition and reduction programs, NIST reduced the uncertainty of air flow calibrations from 0.05 % to 0.025 %. NIST now reduces calibration data for critical flow venturis using the "real" critical flow factor generated by REFPROP v. 9 instead of approximate "ideal" critical flow factors. This change improved the mutual consistency of critical flow venturi calibrations performed in N_2 and air from > 0.05 % to < 0.012 %. In anticipation of future calibrations at higher pressures, we tabulate estimates of property uncertainties for pressures up to 70 MPa for air, argon, helium, N_2, CO_2, H_2 and CH_4 by comparing REFPROP to primary data sources.

Keywords: Properties, gas flow, density, viscosity.

1. Introduction

REFPROP [1] is a NIST-generated and NIST-maintained computer program that calculates thermodynamic and transport properties of pure fluids and mixtures (*e.g.* air, natural gas) at values of the temperature, pressure, and composition specified by the user. REFPROP's authors have collected and critically assessed data from publications of experimental property data, removed outlying data sets, and fit the culled data using the best available models. REFPROP provides properties through a user interface designed for Windows. It also provides compiled FORTRAN code that can be used within other programs (e.g. Excel, Labview) via a dynamic link library (dll). New fluids, new models, and improved property values are added as new versions of REFPROP are released. Version 9 was released in November 2010. It has two new features that are of particular interest to the gas flow measurement community: it calculates 1) the real critical flow factor and 2) transport properties for mixtures that include up to 2% water vapor (*e.g.* moist air).

The most commonly needed properties in gas flow measurement are density (ρ and consequently molecular mass $\mathcal{M}$ and compressibility Z), dynamic viscosity μ, and the critical flow factor C^*.

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[*] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

For example, the density is used for conversion between mass flow and volumetric flow; the viscosity is used to calculate Reynolds number or the Hagen-Poiseuille flow through a laminar flowmeter; and the critical flow factor is used to calculate the theoretical flow through a critical flow venturi.

In this publication we examine version 9 of the NIST properties database REFPROP for the conditions encountered during calibrations by the NIST Fluid Metrology Group (FMG). We undertake the review for three reasons:

1) In January 2009 the FMG changed its property calculations from unpublished fits to REFPROP v. 7 [2] to the new REFPROP dynamic link library. If this change is not considered when customers' meters are recalibrated, it will appear as calibration shifts in customers' control charts. To assist customers, one portion of this paper quantifies the differences between the old and new property results over the range of conditions encountered during normal customer calibrations.

2) The uncertainty of gas properties is needed for flow measurement uncertainty analyses. REFPROP gives uncertainty estimates for many property outputs under the menu item "Substance", "Fluid Information". But the REFPROP uncertainties are specified for a broad range of pressure and temperature; hence these uncertainties are larger than necessary for the more limited conditions used in the FMG. So, another portion of this paper assesses the uncertainty of the properties over the particular range of conditions used within the NIST Fluid Metrology Group.

3) Finally, it is highly desirable to use a single set of property values throughout the entire flow measurement community. We conclude that REFPROP is an accurate and convenient source of such values. Flow calibration laboratories present their results as dimensionless quantities. For instance, in the case of a critical flow venturi, calibration results are a set of values of the discharge coefficient as a function of the Reynolds number. Calculation of these quantities requires molecular mass, viscosity, and the critical flow factor. If a calibration laboratory and the flowmeter user use different values of these properties, the user's CFV flow measurements will have errors. If the lab and user use the same property values, the errors in the property values themselves are correlated and nearly cancel out of the user's flow measurements.[*] Disagreements between participants in inter-laboratory comparisons have been traced to the use of different property values. [3]

2. Labview Program for Gas Properties

NIST's calibration service uses a Labview program called "Gas Properties 9.vi" that uses the REFPROP.dll file to calculate density, viscosity, and C_R^* as well as molecular mass, compressibility, molar density, specific heats, and specific heat ratio for nitrogen, argon, helium, carbon dioxide, hydrogen, methane, and air for a given dew point temperature (K) and pressure at the hygrometer mirror (kPa). The program uses the Labview function "Call Library Function Node" to access the SETUPdll, WMOLdll, TPRHOdll, THERMdll, CSTARdll, and TRNPRPdll functions in the REFPROP.dll. For pure gas calculations these functions are used in a

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[*] The cancellation of errors in property values is not perfect when flow meter calibrations are transferred. For example, if a flowmeter is calibrated at one temperature and pressure and subsequently used at another temperature or pressure, the errors in the property values will differ at the calibration and the use conditions; therefore, they will not perfectly cancel each other.

straightforward manner and the program can be easily extended to other pure gases that are covered by REFPROP by adding new cases to a case structure.

Property calculations for air are more complex in order to account for the presence of water. SETUPdll is run with all of the mixture components listed in Table 1. The air dew point temperature and the pressure of the dew point measurement are used to calculate the partial pressure and mole fraction of water x_{H2O} in the air (corrected by the enhancement factor) [4]. The mole fractions of all dry components in Table 1 are reduced by the factor $(1-x_{H2O})$ and water is added to the array of mole fractions. Then the molecular mass of the moist air is calculated using the mole fractions that include water as input to WMOLdll. This set of constituents and composition are also used to calculate the density, C_R^*, and all other property outputs using TPRHOdll, THERMdll, CSTARdll, and TRNPRPdll. Note that REFPROP 9 is intended for use for mixtures with $x_{H2O} < 0.02$ (dew point temperature < 291 K).

Table 1. Molecular masses and mole fractions for dry air currently used (2010) for flow calibrations.

Component	Molecular Mass (g/mol)	Mole Fraction (2010)
Nitrogen	28.01348	0.780868
Oxygen	31.9988	0.209409
Argon	39.948	0.009332
Carbon Dioxide	44.0098	385×10^{-6}
Helium	4.0026	5×10^{-6}
Water	18.015268	0
	Average Molecular Mass	28.96543

Neon is not included in Table 1, even though its mole fraction in air (18×10^{-6}) is greater than the helium mole fraction in air because REFPROP 9 does not include mixing models for neon. The effects of neglecting neon are negligible ($< 10 \times 10^{-6}$) for the properties of interest herein. The NIST Fluid Metrology Group uses 8.314472 J·mol^{-1}·K^{-1} for the value of the universal gas constant R.

3. Property Differences for 100 kPa to 800 kPa and 270 K to 330 K

In a prior document, I compared the property values used by NIST's Fluid Metrology Group between 2003 and January 2009 with older property calculations. [2] The 2003 property values came from REFPROP v. 7.0 and the comparisons spanned the range of conditions encountered in normal calibrations done in the Fluid Metrology Group: 100 kPa to 800 kPa (in 100 kPa increments) and 270 K to 330 K (in 10 K increments). Here, I compare the v. 9 REFPROP.dll properties calculated via the Labview program Gas Properties 9.vi with the v. 7.0 properties over the same range of conditions.

3.1. N_2, Ar, He, and CO_2

Figures 1 through 4 show the relative differences in density, viscosity, and critical flow factor between the two versions of property calculations. Each relative difference is multiplied by 10^6 and labeled "ppm". At each pressure, seven scaled differences are plotted, one at each of the 10 K increments in the range 270 K $\leq T \leq$ 330 K. The black curve shows the property differences for room temperature conditions (295 K). The density differences are all less than 20 ppm. Viscosity differences are less than 30 ppm for CO_2 and He, but nearly 1400 ppm (0.14 %) for N_2 and nearly 2900 ppm (0.29 %) for Ar. The changes in N_2 and Ar viscosity are expected since REFPROP version 7 was updated based on improved data for transport properties as detailed in reference [5]. We expect significant differences in the viscosity of air for the same reason.
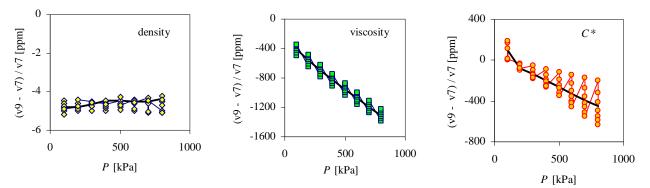
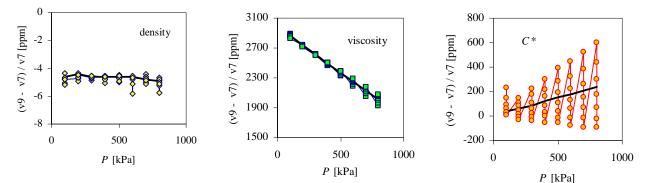*Figure 1. Relative differences between v. 9 and v. 7.0 properties for N_2 at 7 temperatures.*

Figure 2. Relative differences between v. 9 and v. 7.0 properties for Ar at 7 temperatures.
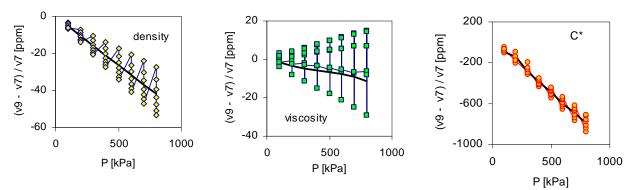
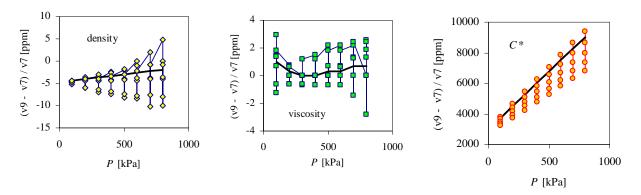*Figure 3. Relative differences between v. 9 and v. 7.0 properties for He at 7 temperatures.*

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Figure 4. Relative differences between v. 9 and v. 7.0 properties for CO_2 at 7 temperatures.

The critical flow factor, C^* is a gas property that NIST uses to correlate calibration data for a critical flow venturi (CFV), one of the most common flowmeter types calibrated at NIST. The physical model for the flow through a CFV requires a relation between the density and speed of sound at the throat of CFV to the stagnation pressure and temperature measured upstream from the CFV. Before REFPROP v. 9, NIST's Fluid Metrology Group used the "ideal critical flow factor", C_I^* calculated from the formula:

$$C_I^* = \sqrt{\gamma \left(\frac{2}{\gamma+1} \right)^{\frac{\gamma+1}{\gamma-1}}} \qquad (1)$$

where $\gamma \equiv C_p/C_v$ is the specific heat ratio that was calculated from polynomial fits to REFPROP v. 7 as documented in reference [2]. As previously stated, REFPROP v. 9 calculates the "real" critical flow factor, C_R^* whereas previous versions did not calculate C^* at all. The definition of the real critical flow factor is,

$$C_R^* = \frac{\rho^* a^* \sqrt{RT_0}}{P_0 \sqrt{\mathcal{M}}} \qquad (2)$$

where ρ^* is the gas density at the nozzle throat, a^* is the sound velocity at the throat, R is the universal gas constant, T_0 is the stagnation temperature, P_0 is the stagnation pressure, and $\mathcal{M}$ is the gas molecular mass. To calculate C_R^*, REFPROP assumes isentropic and iso-energetic expansion from the upstream stagnation conditions to the throat and uses an iterative solver along with the gas properties database. Because the real critical flow factor accounts for non-ideal gas behavior that C_I^* neglects, the values for C^* from v. 7.0♦ and v. 9 differ significantly. For N_2, Ar, and He the relative differences reach 1000 ppm (0.1%); for CO_2 the differences approach 1%.

The change in C^* calculations significantly improved the agreement between CFV calibrations performed in different gases. In reference [6], the agreement of NIST CFV calibrations in air and

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♦ Note that REFPROP v. 7.0 does not actually calculate C^* and what we refer to here as v. 7.0 of C^* is C_I^* based on REFPROP v. 7.0 values of the specific heat ratio.

nitrogen is shown to improve from > 500 ppm to < 120 ppm because of the improved property calculations. The CSTARdll will not give values for CO_2 when input temperatures are < 284 K. REFPROP delivers the message: "[CSTAR error 201]: final state may be 2-phase".

3.2. Property Differences for Air

Relative property differences for dry air are shown in Figure 5. The relative differences in density (< 30 ppm) result from the change of the air composition used in 2003 for the v. 7.0 fits and the composition given in Table 1. The composition change leads to differences in compressibility (pressure and temperature dependent) and an increase in molecular mass of dry air of 28 ppm (28.96464 g/mol in 2003, 28.96543 g/mol in 2010). The viscosity changes range from −670 ppm to −90 ppm and can be traced to REFPROP improvements as explained in reference [5]. The change in C^* is < 350 ppm.
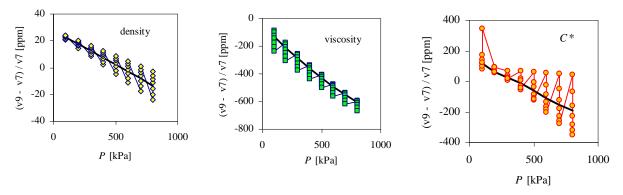
Figure 5. Relative differences between v. 9 and v. 7.0 properties for dry air.

NIST's air calibrations use a compressor and a dryer that discharges air with a typical dew point temperature of −16.7 °C. The 2003 uncertainty analysis for the NIST 34 L and 677 L *PVTt* standards assumed that the air was perfectly dry. The uncertainty analysis treated the difference between the actual, moist air and dry air as an uncertainty component. The addition of an optical hygrometer and REFPROP.dll property calculations reduced the uncertainty of air mass flow measurements in these standards from 0.05% to 0.025% [6].

Figure 6 shows the relative differences in properties between the v. 9 moist air calculation with the typical FMG dew point temperature of −16.7 °C and the fits to v. 7.0 for dry air. The gas is approximately 490 ppm less dense due to the presence of 1400 ppm of water which reduces the air molecular mass to 28.95014 g/mol. The change in viscosity ranges from −470 ppm to −960 ppm and C^* differs from dry air by −360 ppm to 320 ppm.
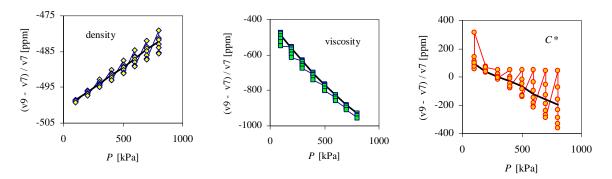
Figure 6. Relative differences between v. 9 and v. 7.0 properties for air with dew point temperature of -16.7 °C corresponding to our typical mole fraction, $x(H_2O) = 0.0014$.

3.3. Viscosity of Moist Air

Viscosity calculations for mixtures that include water vapor up to a mole fraction of 2 % (dew point temperature of 291 K) are a new feature of REFPROP v. 9. Figure 7 compares the viscosity values for moist air from REFPROP v. 9 to 1) viscosities from REFPROP v. 7.0 (dry air viscosity) and 2) calculations following Herning and Zipperer [7]. They use a simpler version of Wilke's [8] formula and the viscosity of the pure components of air (including water) calculated from collision integrals [9]. Unfortunately, the collision integrals do not capture the pressure dependencies of viscosity; they are functions of temperature alone. The largest difference between the three viscosity curves in Figure 7 is 0.5 %.
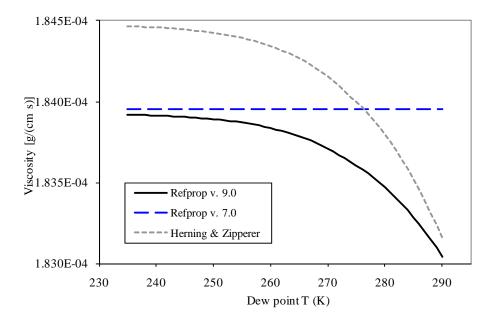

Figure 7. Moist air viscosity from REFPROP v. 9 and two other calculation methods at a pressure of 101 kPa and dew point temperatures from 235 K to 290 K.

4. Effect of Property Changes on Calibration Outputs

The change from REFPROP v. 7.0 to v. 9 will introduce changes in our customers' flowmeter calibration results that could be misinterpreted as shifts in the flowmeters. In this section, we quantify the changes for the two most common flowmeter types calibrated by the Fluid Metrology Group, laminar flowmeters (LF) and critical flow venturis (CFV) when calibrated in air.

The calibration outputs for the laminar flowmeter are the viscosity coefficient and the flow coefficient:

$$VC = \frac{D^2 \rho \, \Delta P}{\mu^2}, \tag{3}$$

$$FC = \frac{D^3 \Delta P}{\mu \, \dot{V}}, \tag{4}$$

where D is a length scale for the LF, ρ is the gas density at the LF, ΔP is the differential pressure, μ is the absolute viscosity, and $\dot{V}$ is the volumetric flow at the LF. To assess the change in the VC and FC introduced by property changes, we note that VC is proportional to ρ / μ^2 and FC to $1/\mu$. We can read the relative density and relative viscosity changes from Figure 6 for our typical LF pressure conditions of 100 kPa: $\Delta\rho_r = -500$ ppm, $\Delta\mu_r = -490$ ppm. Hence the change in VC for typical customer calibrations will be

$$\Delta VC = \Delta\rho_r - 2\Delta\mu_r = 480 \text{ ppm} \tag{5}$$

and the change in FC will be

$$\Delta FC = -\Delta\mu_r = 490 \text{ ppm}. \tag{6}$$

For CFVs, the reported calibration outputs are Reynolds number, Re and discharge coefficient C_d:

$$Re = \frac{4\dot{m}}{\pi \, d \, \mu}, \tag{7}$$

$$C_d = \frac{4 \, \dot{m}\sqrt{RT_0}}{P_0 \pi \, d^2 \, C^* \sqrt{\mathcal{M}}}, \tag{8}$$

where $\dot{m}$ is the mass flow according to a NIST flow standard, d is the CFV throat diameter, and the other variables were defined above in connection with Equation 2. NIST uses *PVTt* standards as the flow reference for most CFV calibrations. A *PVTt* flow standard calculates flow by measuring the density change in a tank of known volume V_T over a time interval t. For present purposes the *PVTt* flow equation is $\dot{m} = \rho_f \, V_T / t$, where ρ_f is the gas density of the full *PVTt* tank at 100 kPa. After substitution of this mass flow equation into Equations 7 and 8, one finds that Re is proportional to ρ/μ and C_d to $\rho / C^* \sqrt{\mathcal{M}}$. From Figure 6, for the full *PVTt* tank pressure (100 kPa), $\Delta\rho_r = -500$ ppm. Unlike the LF case, the pressure at the CFV for a normal

calibration varies and therefore $\Delta\mu_r$ varies between −470 ppm and −960 ppm. Hence the relative change in *Re*,

$$\Delta Re_r = \Delta\rho_r - \Delta\mu_r \qquad (9)$$

ranges between −20 ppm and 470 ppm, depending on the pressure at the CFV. The change in C_d can be evaluated using the formula

$$\Delta C_{d\,r} = \Delta\rho_r - \Delta C_r^* - \frac{1}{2}\Delta\mathcal{M}_r \qquad (10)$$

The relative change in molecular mass $\Delta\mathcal{M}_r$ between the previously used dry air composition and the moist air composition used in our calibrations is −500 ppm. The value of ΔC_r^* varies with the pressure at the CFV and for 290 K ranges between 100 ppm and −200 ppm. Hence the relative change in C_d due to the change in property calculations ranges between −340 ppm at 100 kPa and −40 ppm at 800 kPa.

5. Property Uncertainties for 270 K to 330 K and Various Pressure Ranges

We assessed the uncertainties of density and viscosity values from REFPROP v. 9 for temperatures between 270 K and 330 K in three pressure ranges: 0 MPa to 1 MPa, 0 MPa to 30 MPa, and 0 MPa to 70 MPa. The results are shown in Table 2. The first range spans the conditions of gas flow calibrations normally performed by the NIST Fluid Metrology Group. The other two ranges span conditions that we anticipate adding to our gas flow calibration capabilities in the future.

For most fluids, the REFPROP "Fluid Information" windows give uncertainties for some of the properties REFPROP outputs. Density and viscosity are two of the properties that REFPROP most regularly reports uncertainties for. But the uncertainties are sometimes for larger ranges of conditions than those of interest for the Fluid Metrology Group, so property uncertainties were estimated by studying residual plots available in the literature references listed in the "Fluid Information" windows within REFPROP. Researchers have collected experimental data gathered over many years, qualified it, fit the qualified data, and then plotted the residuals between the fit and the experimental data. The residual plots show the agreement between the experimental measurements of numerous researchers and how well the fits match the experimental data. In general, uncertainty values bounding 95% of the residuals were visually estimated from the literature figures. In a few cases, the literature gave "tolerance diagrams", i.e. regions of pressure and temperature labeled with uncertainty values. Other thermophysical papers give uncertainty estimates in tables listing the experimental data being fitted and these are useful guides for the uncertainty of REFPROP. In cases where no information about property uncertainty was available, the appropriate cell in the table is left blank.

Table 2. The k = 2 (95 % confidence level) uncertainty of density, viscosity, and sound speed for pressures < 1 MPa, < 30 MPa, and < 70 MPa in percent.

	Air	**N$_2$**	**Ar**	**CO$_2$§**	**He**	**H$_2$**	**CH$_4$**
$U(\rho)$	[10] Fig 10	[11] Fig 19	[12] Fig 17	[13] Fig 24	[14] Table 2	[15] Fig 5	[16] Fig 19 & 20
<1 MPa	0.03	0.01	0.01	0.02	0.1	0.02	0.02
<30 MPa	0.03	0.02	0.02	-	0.1	0.04	0.03
<70 MPa	0.03	0.02	0.03	-	0.1	0.06	0.03
$U(\mu)$	[17]	[17]	[17]	[18] Fig 10	[19] Fig 6	[20] Fig 8	[1]
<1 MPa	0.5	0.5	0.5	0.5	0.5	1	0.3
<30 MPa	1	1	1	-	4	2	0.3
<70 MPa	2	1	1	-	-	3	2
$U(a)$	[10] Fig 14	[11] Fig 25	[12] Fig 22	[13] Fig 39	[14]	[21] Fig 29	[16] Table 24 & Fig 22
<1 MPa	0.2	0.02	0.02	0.03	0.1	0.3	0.02
<30 MPa	0.2	0.02	-	-	0.1	0.4	0.05
<70 MPa	-	0.5	-	-	0.1	0.5	0.05

§Large uncertainties occur near the critical point of CO$_2$ (~304 K, ~7.4 MPa) and flow calibrations are not conducted near these conditions, so uncertainties are omitted.

Unfortunately, C_R^* is not directly measured; therefore estimates of its uncertainty do not appear in the literature of thermophysical properties. We plan to estimate the uncertainty of C^* from Equation 2 and the more-readily-available uncertainties of density and air speed. The properties database will be used to generate density and sound speed values that include their uncertainties, i.e. ρ^{*+}, ρ^{*-}, a^{*+}, and a^{*-}. These would be used in the definition of C_R^* to calculate C_R^{*+} and C_R^{*-} and the difference between C_R^{*+} and C_R^{*-} would be used to estimate the uncertainty of C_R^*. In anticipation of this effort, the uncertainty of sound speed has been listed in the following tables.

On occasion, accurate flow calorimetry data are used to constrain model thermodynamic surfaces, such as the ones in REFPROP v. 9. In analogy with this, we foresee that the documented understanding of CFVs will improve and be extended to higher pressures. Eventually, accurate flow measurements will yield values of C_R^* that are so accurate that they to will be used to constrain thermodynamic surfaces.

6. Summary

We assessed the gas properties density, viscosity, and critical flow factor calculated by REFPROP v. 9 over the range of pressures, temperatures, and compositions used for gas flow calibrations in the NIST Fluid Metrology Group. We quantified the change in these properties from the property equations previously used in the FMG (fits to REFPROP v. 7.0) and the uncertainty of the v. 9 outputs.

For the pure gases N_2, Ar, He, CO_2, the relative change in density$^\blacklozenge$ $\Delta\rho_r$ is < 20 ppm. The change in viscosity $\Delta\mu_r$ is < 20 ppm for He and CO_2, but is 0.14 % for N_2 and 0.29 % for Ar due to REFPROP improvements. Changes for the critical flow factor ΔC_r^* are largely due to the differences between the ideal and real versions of this quantity and are approximately 0.1 % for N_2, Ar, and He, and 1 % for CO_2.

Most of the gas flow calibrations performed in the FMG are conducted with air. The addition of an optical hygrometer to measure the dew point temperature of air used in the 34 L and 677 L *PVTt* calibration prompted a revision of the property calculations and a reduction in uncertainty. For air used in the FMG with the typical dew point temperature of −16.7 °C, the relative density change is −490 ppm, $\Delta\mu_r$ is 470 ppm to 960 ppm, and ΔC_r^* is < 360 ppm. The property changes have improved the agreement between calibrations performed in nitrogen and air [6].

We have analyzed the changes that FMG customers can expect in the dimensionless quantities we provide in calibration reports. For laminar flowmeters, the flow coefficient, FC will change by 490 ppm. For critical flow venturis, $\Delta C_{d\,r}$ ranges from −340 ppm at low pressures to 40 ppm at high pressures.

A review of the uncertainties for density, viscosity, and the speed of sound for 270 K to 330 K and three pressure ranges is summarized in Table 2. At low pressures, the uncertainties are often smaller than those given in the REFPROP "Fluid Information" documentation because a more restricted range of pressure and temperature conditions applicable to the FMG was considered.

Acknowledgements

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$^\blacklozenge$ $\Delta\rho = \dfrac{\rho_{v.\,9.0} - \rho_{v.\,7.0}}{\rho_{v.\,7.0}}$

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