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Effect of Refractive Index Variation on Two-Wavelength Interferometry for Fluid Measurements

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Effect of refractive index variation on two-wavelength interferometry for fluid measurements

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Abstract
Two wavelength interferometry can in principle be used to measure changes in both temperature and concentration in a fluid, but measurement errors may be large if the fluid dispersion is small. This paper quantifies the effects of uncertainties in dn/dT and dn/dC on the measured temperature and concentration when using the simple expression dn = (dn/dT)dT + (dn/dC)dC. For the data analyzed here, ammonium chloride in water from -5 to 10 °C over a concentration range of 2 - 14% and for wavelengths 514.5 and 633 nm, it is shown that the gradients must be known to within 0.015% to produce a modest 10% uncertainty in the measured temperature and concentration. These results show that great care must be taken to ensure the accuracy of refractive index gradients when using two wavelength interferometry for the simultaneous measurement of temperature and concentration.

Introduction
Interferometry is an excellent method for measuring refractive index changes in fluids with high accuracy. When the refractive index changes are caused by changes in a single flow variable such as temperature, it is straightforward to convert interferometric data to engineering data. When two properties change the refractive index simultaneously, two-wavelength interferometry has been used to get an extra equation to solve for the additional unknown.[1] Similarly, three wavelengths can be used to solve for three unknowns. There are several sources of error in two wavelength interferometry. Some are common to interferometers in general, such as errors caused by fringe measurement error[2,3] and errors caused by refraction and end effect.[4] However, as mentioned by Vest[5] with regard to gases, two wavelength interferometry is very sensitive to small errors when the fluid is weakly dispersive. This paper presents an uncertainty analysis that quantifies the relationship between calculated temperature and concentration and the values used for dn/dT and dn/dC for the following relationship:[6]

$$\Delta n = \left(dn/dT\right)T + \left(dn/dC\right)C \tag{1}$$

where n is the refractive index, T is the temperature, and C is the concentration of the fluid mixture. Data specific to ammonium chloride in water is then analyzed because of its wide use for simulating superalloy solidification[7] in microgravity studies. It is shown that for ammonium chloride over the temperature range -5 to 10 °C over a concentration range of 2 - 14% and for wavelengths 514.5 and 633 nm the values of dn/dT and dn/dC must be known to high accuracy over the experimental range to ensure reasonable uncertainties in the final measurements of temperature and concentration.

Procedure

Interferometry relates measured interference fringe spacings to refractive index differences as follows:

$$\lambda \frac{\Delta M_\lambda}{\Delta s} = L \frac{\Delta n}{\Delta s}\Big|_\lambda \tag{2}$$

where λ is the wavelength of light, L is the physical pathlength through which the light passes, Δs is a pathlength difference in the plane normal to the line-of-sight, and ΔM and Δn are the number of fringes, and refractive index change along Δs, respectively. The subscripts λ refers to wavelength dependent quantities. The pathlength L is known from the experiment, the wavelength is known by choice of laser operating line, and $\Delta M/\Delta s$ is measured from an interferogram. Knowledge of these parameters permits the calculation of $\Delta n/\Delta s$ for a particular wavelength. Rewriting equation (1) to describe the line-of-sight integration of interferometric measurements yields:

$$\frac{dn}{ds}\Big|_\lambda = \frac{dn}{dT}\Big|_\lambda \frac{dT}{ds} + \frac{dn}{dC}\Big|_\lambda \frac{dC}{ds}. \tag{3}$$

In this case there are two unknowns, dT/ds and dC/ds. Therefore two wavelength interferometry should be sufficient for producing two equations required for a solution. Equations (2) and (3) are combined for each wavelength:

$$\frac{\lambda_i}{L}\frac{dM_i}{ds} = \frac{dn}{dT}\Big|_{\lambda i} \frac{dT}{ds} + \frac{dn}{dC}\Big|_{\lambda i} \frac{dC}{ds} \tag{4}$$

where i=1,2 represents the two wavelengths. Implicit in the assumption of a solution is that dn/dT and dn/dC are different for two different wavelengths, and that:

$$\frac{(dn/dT)\big|_{\lambda 1}}{(dn/dT)\big|_{\lambda 2}} \neq \frac{(dn/dC)\big|_{\lambda 1}}{(dn/dC)\big|_{\lambda 2}} \tag{5}$$

Forming a new variable from the experimentally determined quantities:

$$\Lambda_i = (\lambda_i/L)(dM_i/ds) \tag{6}$$

and shortening the nomenclature by the following new variables:

$$c_{1T} = \frac{dn}{dT}\Big|_{\lambda 1} \qquad c_{2T} = \frac{dn}{dT}\Big|_{\lambda 2}$$

$$c_{1C} = \frac{dn}{dC}\Big|_{\lambda 1} \qquad c_{2C} = \frac{dn}{dC}\Big|_{\lambda 2} \tag{7}$$

yields the simplified equations:

$$\Lambda_1 = c_{1T}(dT/ds) + c_{1C}(dC/ds)$$

$$\Lambda_2 = c_{2T}(dT/ds) + c_{2C}(dC/ds) \tag{8}$$

which can be used to solve for dT/ds and dC/ds:

$$\frac{dT}{ds} = \frac{\Lambda_1 c_{2C} - \Lambda_2 c_{1C}}{c_{1T}c_{2C} - c_{1C}c_{2T}}$$

$$\frac{dC}{ds} = \frac{\Lambda_1 c_{2T} - \Lambda_2 c_{1T}}{c_{1C}c_{2T} - c_{1T}c_{2C}} \tag{9}$$

Note that Equation (5) must be satisfied or neither of these quantities can be determined.

What is the uncertainty in dT/ds and dC/ds (and therefore also in the measured temperature T and concentration C)? Assuming that uncertainties in the variables on the right hand side in Eqn. (9) are independent, uncertainty analysis yields:[8]

$$\varepsilon_{dTds}^2 = \left[\frac{\partial(dT/ds)}{\partial c_{1T}} \varepsilon_{c1T} \right]^2 + \left[\frac{\partial(dT/ds)}{\partial c_{2T}} \varepsilon_{c2T} \right]^2 + \left[\frac{\partial(dT/ds)}{\partial c_{1C}} \varepsilon_{c1C} \right]^2 + \left[\frac{\partial(dT/ds)}{\partial c_{2C}} \varepsilon_{c2C} \right]^2$$

$$\varepsilon_{dCds}^2 = \left[\frac{\partial(dC/ds)}{\partial c_{1T}} \varepsilon_{c1T} \right]^2 + \left[\frac{\partial(dC/ds)}{\partial c_{2T}} \varepsilon_{c2T} \right]^2 + \left[\frac{\partial(dC/ds)}{\partial c_{1C}} \varepsilon_{c1C} \right]^2 + \left[\frac{\partial(dC/ds)}{\partial c_{2C}} \varepsilon_{c2C} \right]^2$$

(10)

where ε_x represents the uncertainty in variable x. Uncertainties caused by the wavelength, geometric pathlength, and number of fringes per unit distance have not been included here. This analysis is meant to determine the effects of the refractive index variations with concentration and temperature only. From Eqn. (9) the partial derivatives can be calculated:

$$\frac{\partial(dT/ds)}{\partial c_{1T}} = \left(\frac{-c_{2C}}{c_{1T}c_{2C} - c_{1C}c_{2T}} \right) \frac{dT}{ds}$$

$$\frac{\partial(dT/ds)}{\partial c_{2T}} = \left(\frac{c_{1C}}{c_{1T}c_{2C} - c_{1C}c_{2T}} \right) \frac{dT}{ds}$$

$$\frac{\partial(dT/ds)}{\partial c_{1C}} = \left(\frac{c_{2T}}{c_{1T}c_{2C} - c_{1C}c_{2T}} - \frac{R\lambda_2/\lambda_1}{c_{2C} - c_{1C}R\lambda_2/\lambda_1} \right) \frac{dT}{ds}$$

$$\frac{\partial(dT/ds)}{\partial c_{2C}} = \left(\frac{c_{2T}}{c_{1T}c_{2C} - c_{1C}c_{2T}} - \frac{R\lambda_2/\lambda_1}{c_{2C} - c_{1C}R\lambda_2/\lambda_1} \right) \left(\frac{-c_{1C}}{c_{2C}} \right) \frac{dT}{ds}$$

(11)

Similarly,

$$\frac{\partial(dC/ds)}{\partial c_{1T}} = \left(\frac{c_{2C}}{c_{1C}c_{2T} - c_{1T}c_{2C}} - \frac{R\lambda_2/\lambda_1}{c_{2T} - c_{1T}R\lambda_2/\lambda_1} \right) \frac{dC}{ds}$$

$$\frac{\partial(dC/ds)}{\partial c_{2T}} = \left(\frac{c_{2C}}{c_{1C}c_{2T} - c_{1T}c_{2C}} - \frac{R\lambda_2/\lambda_1}{c_{2T} - c_{1T}R\lambda_2/\lambda_1} \right) \left(\frac{c_{1T}}{c_{2T}} \right) \frac{dC}{ds}$$

$$\frac{\partial(dC/ds)}{\partial c_{1C}} = \left(\frac{-c_{2T}}{c_{1C}c_{2T} - c_{1T}c_{2C}} \right) \frac{dC}{ds}$$

$$\frac{\partial(dC/ds)}{\partial c_{2C}} = \left(\frac{c_{1T}}{c_{1C}c_{2T} - c_{1T}c_{2C}} \right) \frac{dC}{ds}$$

(12)

where

$$R \equiv \frac{dM_2/ds}{dM_1/ds},$$ (13)

the ratio of the number of interference fringes per unit length measured from wavelength 2 and 1. From all of these equations, we can determine the effect of inaccuracy in dn/dT and dn/dC on the measured concentration and temperature.

The variation of refractive index with temperature and concentration has been determined for two wavelengths 632.8nm and 514.5nm by Spatz.[9] The reported relationship is:

$$n_{HeNe} = 1.3332 - \left(6.8276 \cdot 10^{-5}\right)T + \left(1.6764 \cdot 10^{-3}\right)C$$

$$n_{Ar} = 1.3373 - \left(7.1010 \cdot 10^{-5}\right)T + \left(1.7345 \cdot 10^{-3}\right)C$$ (14)

where temperature T is expressed in °C and concentration C is expressed in % by weight. These equations were stated to be valid for the a temperature range -5 °C to 10 °C and concentrations of 2% to 14% by weight NH_4Cl in water. Spatz also reports a value for R.[9] The required constants are summarized in Table 1.

	Symbol	Value
	R =	1.289
	$\lambda 1$ =	632.8 nm
	$\lambda 2$ =	514.5 nm
dn/dT @ $\lambda 1$	c_{1T} =	6.8276E-5 (°C)$^{-1}$
dn/dT @ $\lambda 2$	c_{2T} =	7.1010E-5 (°C)$^{-1}$
dn/dC @ $\lambda 1$	c_{1C} =	1.6764E-3
dn/dC @ $\lambda 2$	c_{2C} =	1.7345E-3

Table 1.   Values of constants.

Substituting these values into Equation (12) yields the following dependence of dT/ds and dC/ds on errors in the four constants c_{iT} and c_{iC}:

$$\frac{\Delta(dC/ds)}{dC/ds} = 396.8\left(\frac{\Delta c_{1T}}{c_{1T}}\right) = 396.8\left(\frac{\Delta c_{2T}}{c_{2T}}\right)$$

$$\frac{\Delta(dC/ds)}{dC/ds} = -266.5\left(\frac{\Delta c_{1C}}{c_{1C}}\right) = 265.5\left(\frac{\Delta c_{2C}}{c_{2C}}\right)$$ (15)

and

$$\frac{\Delta(dT/ds)}{dT/ds} = 265.5\left(\frac{\Delta c_{1T}}{c_{1T}}\right) = 266.5\left(\frac{\Delta c_{2T}}{c_{2T}}\right)$$

$$\frac{\Delta(dT/ds)}{dT/ds} = -178.3\left(\frac{\Delta c_{1C}}{c_{1C}}\right) = 178.2\left(\frac{\Delta c_{2C}}{c_{2C}}\right)$$ (16)

From Equations (15) and (16) it is clear that small errors in the four constants produce large errors in the calculated temperature and concentration distributions. In particular, a 1% uncertainty in dn/dT for either wavelength produces a nearly 4-fold change in the calculated concentration, and a factor of 2.6 error in temperature. A 1% uncertainty in

dn/dC produces a factor of 2.6 error in the calculated concentration and 180% error in temperature. Note that using a value of dn/dT = 6.853E-5 instead of 6.8276E-5 for λ=632.8nm violates Eqn. (5) and prevents the determination of either concentration or temperature. This change in dn/dT represents a change of only -0.372%. Figure (1) shows n plotted as a function of temperature using both dn/dT = 6.853E-5 and 6.8276E-5 $(°C)^{-1}$ in Equation (14a). The data are practically indistinguishable, certainly within the standard deviation in the data presented in Ref. 9. Yet this small difference produces huge changes in the measured concentration and temperature.

These errors have been considered independently. Equation (10) determines to total uncertainty if each of the four constants can be considered to vary independently with equal weight. Normalizing Equation (10) yields:

$$\left[\frac{\varepsilon_{dTds}}{dT/ds}\right]^2 = \left[\frac{\partial\left(\frac{dT}{ds}\right)}{\partial c_{1T}}c_{1T}\frac{\varepsilon_{c1T}}{c_{1T}}\right]^2 + \left[\frac{\partial\left(\frac{dT}{ds}\right)}{\partial c_{2T}}c_{2T}\frac{\varepsilon_{c2T}}{c_{2T}}\right]^2 + \left[\frac{\partial\left(\frac{dT}{ds}\right)}{\partial c_{1C}}c_{1C}\frac{\varepsilon_{c1C}}{c_{1C}}\right]^2 + \left[\frac{\partial\left(\frac{dT}{ds}\right)}{\partial c_{2C}}c_{2C}\frac{\varepsilon_{c2C}}{c_{2C}}\right]^2$$

$$\left[\frac{\varepsilon_{dCds}}{dC/ds}\right]^2 = \left[\frac{\partial\left(\frac{dC}{ds}\right)}{\partial c_{1T}}c_{1T}\frac{\varepsilon_{c1T}}{c_{1T}}\right]^2 + \left[\frac{\partial\left(\frac{dC}{ds}\right)}{\partial c_{2T}}c_{2T}\frac{\varepsilon_{c2T}}{c_{2T}}\right]^2 + \left[\frac{\partial\left(\frac{dC}{ds}\right)}{\partial c_{1C}}c_{1C}\frac{\varepsilon_{c1C}}{c_{1C}}\right]^2 + \left[\frac{\partial\left(\frac{dC}{ds}\right)}{\partial c_{2C}}c_{2C}\frac{\varepsilon_{c2C}}{c_{2C}}\right]^2$$ (17)

and using the values from Table 1 yields:

$$\left[\frac{\varepsilon_{dTds}}{dT/ds}\right]^2 = \left[2655\frac{\varepsilon_{c1T}}{c_{1T}}\right]^2 + \left[2665\frac{\varepsilon_{c2T}}{c_{2T}}\right]^2 + \left[-178.3\frac{\varepsilon_{c1C}}{c_{1C}}\right]^2 + \left[1.783\frac{\varepsilon_{c2C}}{c_{2C}}\right]^2$$

$$\left[\frac{\varepsilon_{dCds}}{dC/ds}\right]^2 = \left[398.6\frac{\varepsilon_{c1T}}{c_{1T}}\right]^2 + \left[396.8\frac{\varepsilon_{c2T}}{c_{2T}}\right]^2 + \left[-266.5\frac{\varepsilon_{c1C}}{c_{1C}}\right]^2 + \left[2655\frac{\varepsilon_{c2C}}{c_{2C}}\right]^2$$ (18)

Assuming just a 0.1% error in each of the four constants yields:

$$\varepsilon_{dTds} = 0.4529\left(dT/ds\right)$$
$$\varepsilon_{dCds} = 0.6756\left(dC/ds\right)$$ (19)

that is, a 0.1% uncertainty in each refractive index variation produces a 45% uncertainty in temperature and a 67% uncertainty in concentration. These values scale linearly with assumed refractive index gradient uncertainties. To measure temperature and concentration to within 10%, dn/dT and dn/dC must be known to within 0.015%.

Conclusions
Uncertainty analysis was used to determine the accuracy with which dn/dT and dn/dC need to be made to ensure sufficient temperature and concentration accuracy from two-wavelength interferograms. For the data analyzed here, namely a temperature range from -5 to 10 °C and a concentration range of 2% to 14% by weight NH_4Cl in water for 632.8 and 514.5nm wavelengths, and using the simple linear expression relating refractive index to temperature and concentration described by Equation 1, the uncertainty must be

0.015% or less for both dn/dT and dn/dC to measure the temperature and concentration to within 10%. This excludes all other error sources, including fringe measurement errors which can contribute up to 20% error.[2] Values for dn/dT are commonly obtained by using $dn/dT = (-3/2)[n(n^2-1)/(2n^2+1)]\beta$, where β is the coefficient of thermal expansion, but this expression is only accurate to about 2%.[10] Refractive index measurements accurate to four decimal places can be made by placing a liquid in a parallel sided glass cell, focusing a beam first on the front surface of the liquid and then the rear surface and measuring the focus shift.[11] Obtaining this accuracy requires distance measurements on the order of 100 nm and angular measurements on the order of a few arc seconds. Certainly the measurements must be made for the specific fluid under study over the specific temperature and concentration ranges, not extrapolated from published data.

These very large final errors from small errors in dn/dT and dn/dC are the direct consequence of the slow variation of dn/dT and dn/dC with wavelength in ammonium chloride. This behavior is typical of fluids and gases.

Implicit in these results is the assumption of the relationship between refractive index, temperature and concentration given by Equation 1. Recently researchers have refined this relationship to include quadratic terms[12] and fourth-power plus cross terms in the Cauchy equation.[7] This latter expression is complex, including thirty coefficients. Unfortunately, increasing the number of coefficients neither compensates for measurement noise nor increases the dispersion of the fluid and therefore should not reduce the sensitivity of two-wavelength interferometry to uncertainties in refractive index gradients.

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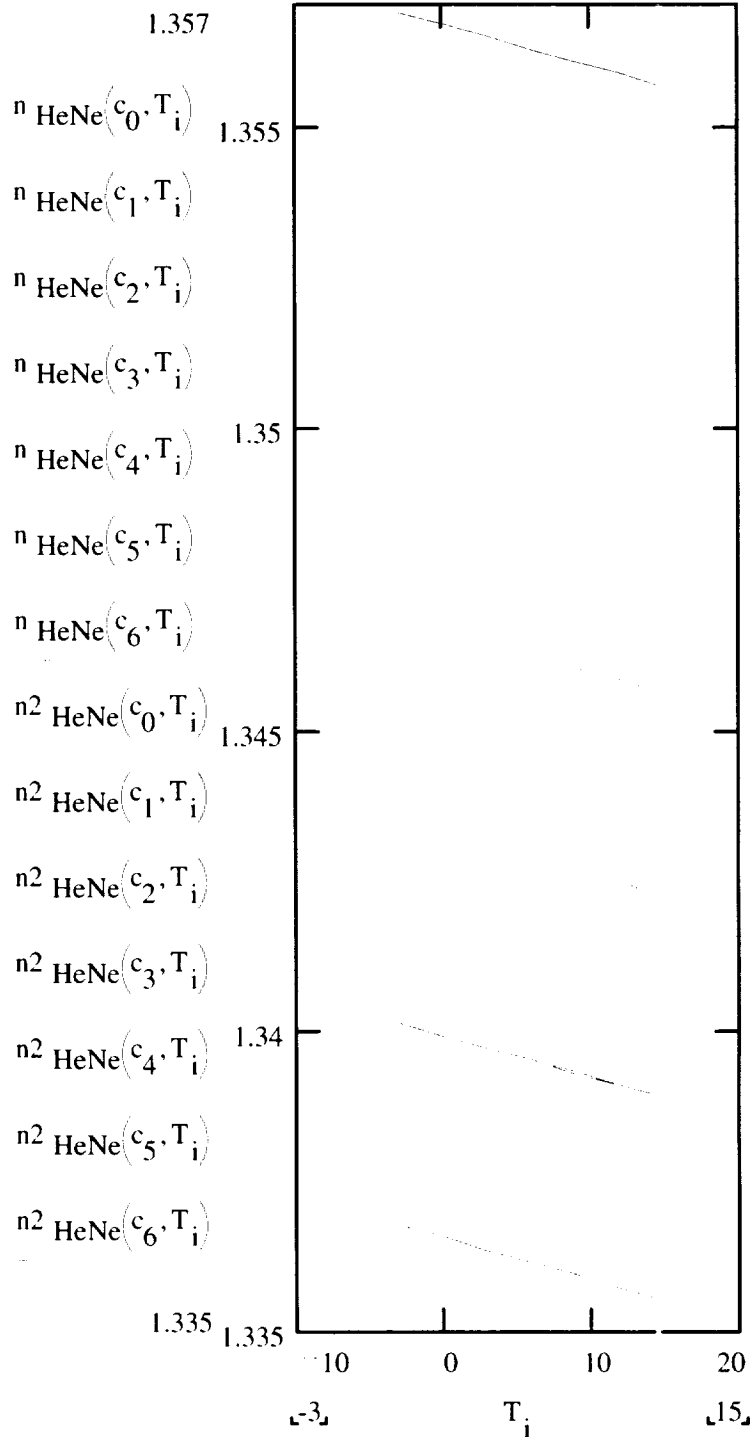
FIGURE 1. Variation of refractive index with temperature for λ = 632.8 nm, concentration ranging from 2% to 14% in 2% intervals, and two values of dn/dT: 6.853E-5 and 6.8276E-5 $(^\circ C)^{-1}$.

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