

# Measurements of Multicomponent Microdroplet Evaporation by Using Novel Optical Techniques

by

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## ABSTRACT

The evaporation process of a multi-component droplet is complex because it involves transient interfacial heat transfer, multi-component phase change and diffusion process within the droplet. Among the difficulties associated with the study of the fluid dynamics and heat transfer of multi-component droplet evaporation are the extremely different volatilities and the rapid variation of the mass fraction within a droplet of the components. Because of the complexity of multi-component droplet evaporation process and the lack of experimental data on the transient evaporation rate, the mass fraction of the components and the droplet interaction effects, the mechanisms controlling the phenomena of multi-component microdroplet evaporation are not well understood. Advanced optical diagnostic instruments have been developed in these experiments for simultaneously measuring the size, velocity and internal temperature of individual spherical evaporating multicomponent microdroplet. A recently improved laser phase-Doppler anemometry system provides high accuracy in the size and velocity measurements. The temperature of a microdroplet was measured utilizing primary rainbow refractometer. Single- and multi-component microdroplets were used in the experiments. Evaporation of multicomponent microdroplets was studied utilizing novel phase-Doppler anemometry and rainbow refractometer. Single- and multi-component microdroplets were used in the experiments. Various droplet diameters, velocities and channel air temperatures were used in the experiments. With the newly developed optical diagnostic method, it is possible to probe the evaporation rate of a multi-component droplet. It was found that the droplet temperature could hardly reach homogenous within the droplet unlike explained by previous researchers. This finding can be explained using the temperature gradient within the evaporating microdroplets. Due to inhomogeneous temperature distribution and the evaporation at the liquid vapor interface the measured temperature by a rainbow refractometer neither represent the actual temperature of the droplet nor the mean temperature of it. The effect of temperature gradient within an evaporating microdroplet has been investigated.

## INTRODUCTION

Mass and energy exchange between a dispersed phase and an ambient gas in multiphase flows is encountered in numerous atmospheric and industrial processes. Such flows cover a wide range of applications including spray cooling, spray combustion, spray fire suppression, spray drying and air-fuel premixing in combustors. All of these situations involve complex nonlinear couplings of momentum, energy, and mass exchange. The evaporation process of multicomponent droplets is complex because it involves transient interfacial heat transfer, multicomponent phase change and diffusion process within the droplets. Among the difficulties associated with the study of the fluid dynamics and heat transfer of multicomponent droplet evaporation are the extremely different volatilities and the rapid variation of the mass fraction within a droplet of the components. Because of the complexity of multicomponent droplet evaporation process and the lack of experimental data on the transient evaporation rate, the mass fraction of the components and the droplet interaction effects, the mechanisms controlling the phenomena of multicomponent micro-drop evaporation and spray deposition, are not well understood.

The progress in understanding the behavior of droplets in temperature field has relied on experimental investigation due to the complexity of the evaporation process. Such quantitatively experimental studies are rare even for single component sprays because of the difficulties involved in performing reliable experiments on sprays evaporating in a heated gas flow under well defined boundary conditions. The evaporation of a Freon-11 spray injected into stagnant air was studied by Solomon et al. (1985) by using rather old fashioned methods, such as isokinetic sampling technique for droplet concentration and photographic and impacting methods for droplet size distribution. Furthermore, they obtained no correlation between droplet size and velocity. More detailed measurements of a non-evaporative spray interacting with a co-flow turbulent air stream were performed by Rodoff et al. (1987) using laser phase-Doppler anemometry. However, neither heat transfer nor evaporation process was conducted in their study. Experimental studies of a vaporizing single component spray issuing into a co-flow turbulent air stream were performed by Hanson (1952), Yule et al. (1983). Again, no quantitative measurements of droplet velocity and size, density, temperature and species evaporation rate were available for multicomponent droplet evaporation. Sommerfeld et al (1993) and Sommerfeld and Qiu (1997) studied one component spray evaporation under well defined boundary conditions by using a recently developed method for droplet mass flux measurements. Yao and Choi (1987) developed a mono-dispersed spray generator, which allowed the liquid mass flux, droplet size and droplet velocity of the spray to vary independently. It is only recently that much advances have been achieved when phase-Doppler anemometry (PDA) becomes available (Durst and Zaré, 1975). In the past few years, advanced optical diagnostic instruments have been developed in these experiments for simultaneously measuring the size, velocity and internal temperature gradient of individual spherical multicomponent microdroplet. A recently improved laser phase-Doppler anemometry system provides high accuracy in the size and velocity measurements (Qiu et al 1999). The temperature of a microdroplet was measured utilizing primary rainbow refractometer (Roth et al (1991), Sankar et al (1993)). Single- and multi-component microdroplets were used in the experiments. Various droplet diameters, velocities and channel air temperatures were used in the experiments. With the newly developed optical diagnostic method, it is possible to probe the evaporation rate of a multi-component droplet.

In this study multicomponent microdroplet evaporation process has been investigated utilizing novel phase-Doppler anemometry and rainbow refractometer. The problem encountered in the temperature gradient within a droplet using rainbow refractometer will be discussed in this paper. The flow effects have also be investigated. Analysis based on geometric optics method dealing with light scattering from a non-homogeneous sphere and its impact on the measurements using rainbow refractometer and phase-Doppler anemometry has been conducted. A correction method for reducing the measurement errors will be proposed. The measurement techniques including the optical probing technique, particle sizing and experimental data will be described in details in this paper.

## EXPERIMENTAL SETUP

Figure 1 shows the schematic of the experimental setup. The heating system was designed and manufactured utilizing hot silicon oil circulation method. Because this system should cooperate with the droplet generator, the PDA system and the rainbow refractometer, a cylindrical quartz central-channel furnace was fabricated to make the furnace transparent. The channel temperature was adjusted by a U-shape heater which was immersed in the silicon oil and controlled by an electric controller. Using quartz glass is because of its excellent thermal and optical properties. Thermocouples were used to measure the temperature of silicone oil and hot air in the central-channel furnace. As a result, the temperature of the hot air along the axis of the central channel was controlled. To reduce the temperature gradient of the silicone oil in the beaker and to heat the air in the central channel more effectively, an electrical stirrer was installed in the silicone oil.

To reduce heat loss, the Teflon cover, base and combined adapter was applied, which also prevented the hot air escape from the top of central channel. As Teflon is a good heat insulation material, heat expansion is very low even under high temperature conditions. The U-shaped heater, electrical stirrer and thermocouples were arranged to avoid any obstruction of the laser beams both in the transmitting and scattering sides as shown in Figure 2. The channel temperature can vary from the ambient temperature to 200°C. The vibrating orifice mono-dispersed droplet generator (Model 3450, TSI Inc.) was used to generating multicomponent microdroplets for the experiments. Microdroplets passed the channel vertically along the axis of the channel.

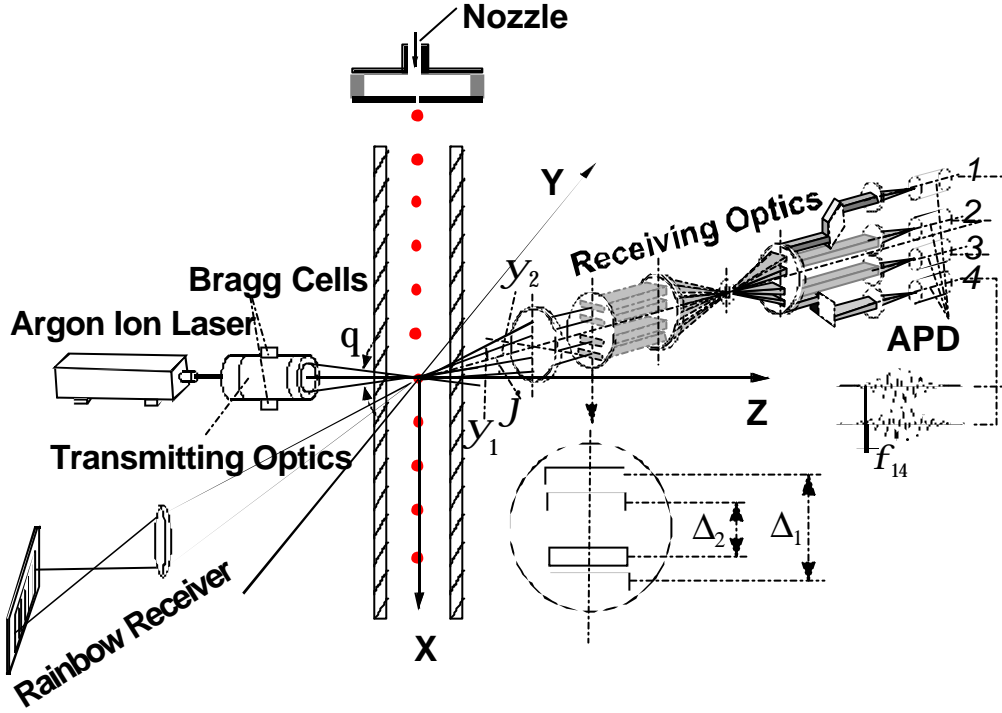


Figure 1 Schematic of Experimental Setup

To measure the droplet velocity and diameter during the evaporation process, a previously developed novel PDA system was used as shown in Figure 1. To reduce the measurement volume effect, the recently improved 4-detector PDA system consists of four detectors that are symmetrically orientated with elevation angles  $y_1$  and  $y_2$  for outer and inner detector pairs, respectively. The phase differences  $f_{14}$  and  $f_{23}$  between the two outer and inner detectors can be determined from the two Doppler signal pairs. According to previous analysis, the optimized receiving angle  $j$  can be determined from:

$$j = \arccos \left[ \left( \frac{1}{4m^2} \left( 1 + 4m^2 + \sqrt{1 + 8m^2} \right) - 1 \right) \right] \quad (1)$$

where  $m$  is the initial refractive index of liquid before heated. Therefore, the relationship between the measured phases from multi-detectors and optical parameters can be described in the following equation.

$$\sin \left( \frac{f_{14} + C_{12} D}{2} \right) \sin \left( \frac{f_{23} - C_{20} D}{2} \right) = \sin \left( \frac{f_{14} - C_{10} D}{2} \right) \sin \left( \frac{f_{23} + C_{22} D}{2} \right) \quad (2)$$

where  $C_2$  and  $C_0$  are the conversion factors of second order refraction from negative side and the surface reflection, which can be calculated from the optical geometry as shown in Figure 1. The only unknowns are the droplet diameter,  $D$ , and the refractive index of the droplet. Therefore, if, the refractive index of the droplet can be measured, the droplet diameter can be determined if  $f_{14}$  and  $f_{23}$  can be measured.

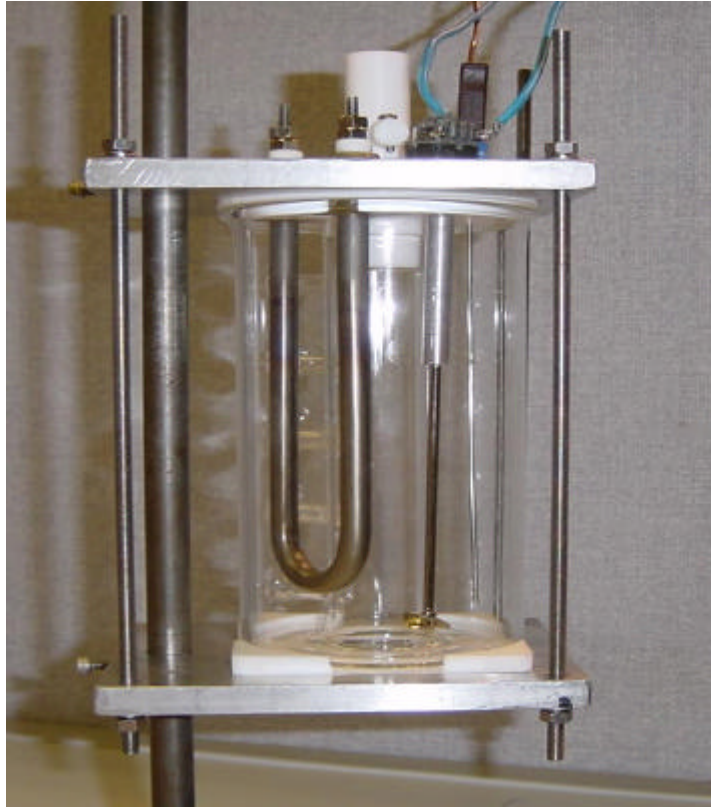


Figure 2 The experimental heating channel

To determine the droplet refractive index, a rainbow refractometer was used and the position of the primary rainbow angle was orientated. The single blue beam (488.0nm) from the Argon Ion laser was used for rainbow refractometer in the experiments. Because rainbow rays exhibit an inherent tendency to be strongly polarized perpendicular to the scattering plane, the blue beam was polarized perpendicular to the plane containing the optical transmitter and optical receiver. The schematic of the rainbow refractometer is presented in Figure 1 which consists of a high resolution CCD camera (Model DRC-TRV 900E, Sony) with a Lens (micro Nikkor 105mm f/2.8, Nikon) and a computer system to record the image of the rainbow. Different test liquids used include water ( $m=1.3336$ ), pure ethanol ( $m=1.36$ ), and different components of ethanol. At room temperature, linearity of the rainbow angle with refractive index could be established; linearity of the CCD pixel values with the scattering angle could also be established. So the location of CCD camera will be calibrated. Using the computer software (Capview) the fringe images of rainbow can be recorded. Because the rainbow angle is known to vary with the droplet size, a refractive index histogram was generated for each size class. The peak location of the histogram was then assumed to be the measured rainbow location for that particular size class.

The droplet temperature can be determined by the measured refractive indexes of droplets according to the calibration curves shown in

Figure 3. The temperature of the heat system was initially set to 21 degree Celsius. Pure water droplets as testing liquid were generated. By adjusting the frequency of the generator 71um in diameter droplets can be generated. At the inlet of the cylindrical central-channel, the droplet diameter, velocity and refractive index were measured by PDA system and the rainbow refractometer. Then, let the water droplet pass through the cylindrical central-channel (165mm) and the droplets were evaluated at different distances downstream of the nozzle as well as the outlet of the channel. In order to reduce the error of the operation, several images measured by sequential monodispersed droplets were recorded to get the mean primary rainbow angle pixel value later.

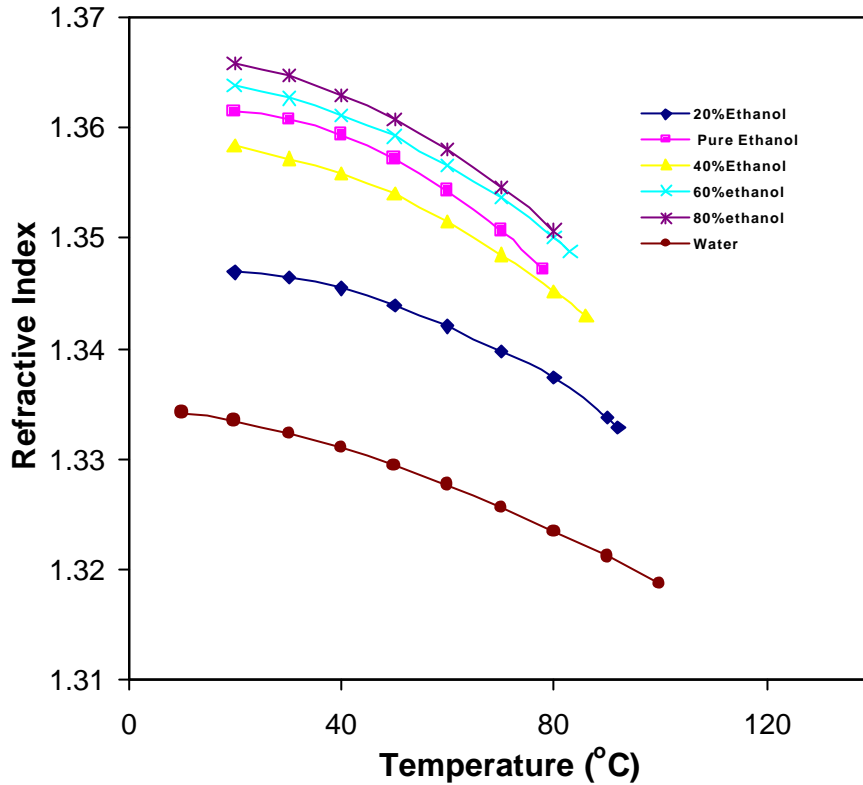


Figure 3 The relationship between the temperature and the refractive index for different test liquids

Increasing the temperature of the heat system, each time increased 10 degree Celsius, until the temperature reached to 200 degree Celsius. Repeated the steps above, the results of water can be obtained. 20%, 40%, 60%, 80% ethanol and pure ethanol were also measured following the steps above for water. As the refractive index of ethanol with different percent is different, when using PDA system to test the size of droplet, the different off-axis angles for optical parameters of PDA system were set. For example, as 20% ethanol, refractive index set 1.34,  $\phi=47^\circ$  according to Eq. (1).

## RESULTS AND DISCUSSION

The fringe images of water in different temperature were shown in Figure 4 with which the droplet temperature can be calculated.

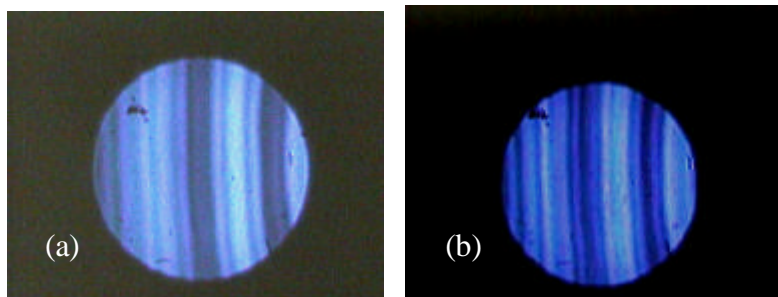


Figure 4 The rainbow fringe images with different temperatures for water (a)  $T_a=21^\circ\text{C}$ , (b)  $T_a=200^\circ\text{C}$

Figure 5 shows the evaporating water droplet diameter as a function of hot air temperature. In this case, the droplets were measured at the immediate outlet of the channel. The linear relationship between the hot air temperature and the droplet diameter can be assumed.

Figure 6 shows the water droplet temperature as the function of hot air temperature. The measurement location was the same as indicated in Figure 5. From Figure 6, it can be seen that the measured maximum water droplet temperature is always below the boiling point temperature of water. This might be explained as because the measured droplet temperature is neither the surface temperature nor the average temperature of the droplet when a temperature gradient is existed within the droplet, which is true in most cases, the measured temperature is always the temperature between the surface temperature (the boiling temperature) and the temperature of its centre location. However, Figure 6 also indicated that the droplet temperature has never reached homogenous within the droplet unlike explained by previous researchers.

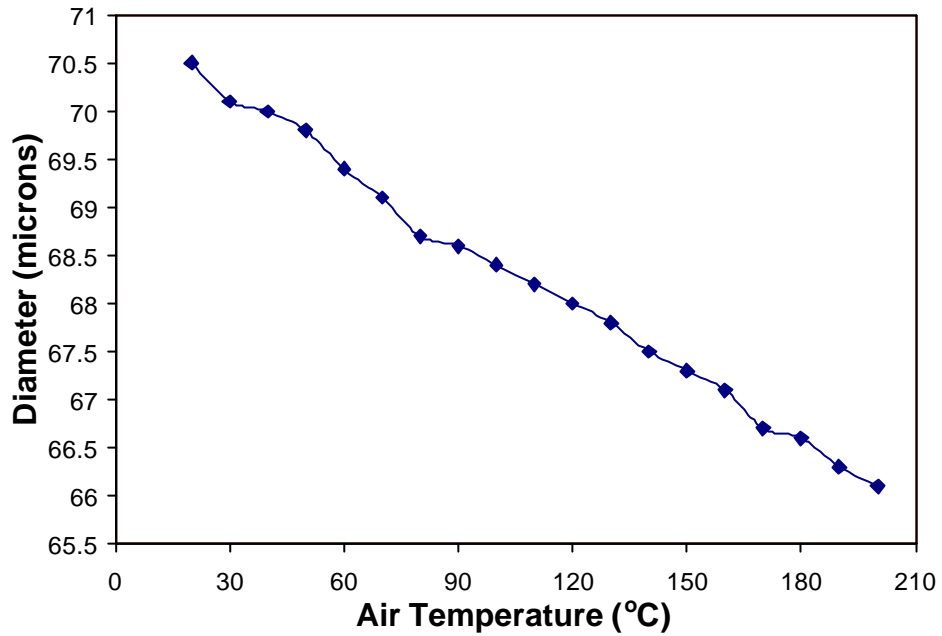


Figure 5 The measured water droplet diameter via air temperature field

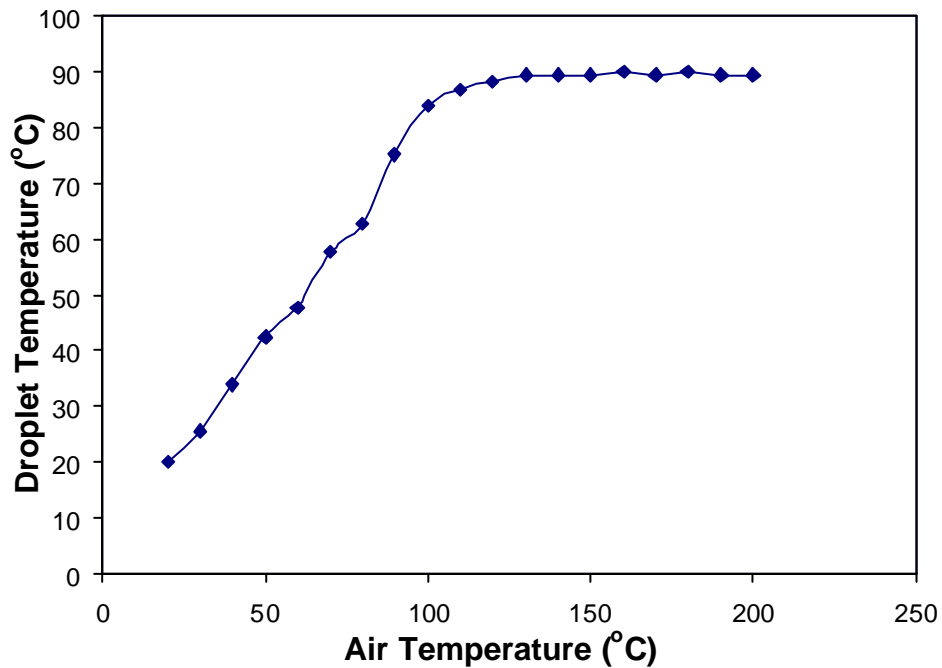


Figure 6 The measured water droplet temperature via the hot air temperature field

Figure 7 shows the measured rainbow fringe pattern for 20% ethanol. To convert the measured refractive index to temperature, the calibration curve of refractive index via temperature for 20% ethanol was adopted as shown in Figure 8 (N.C. Thorlen, 1988).

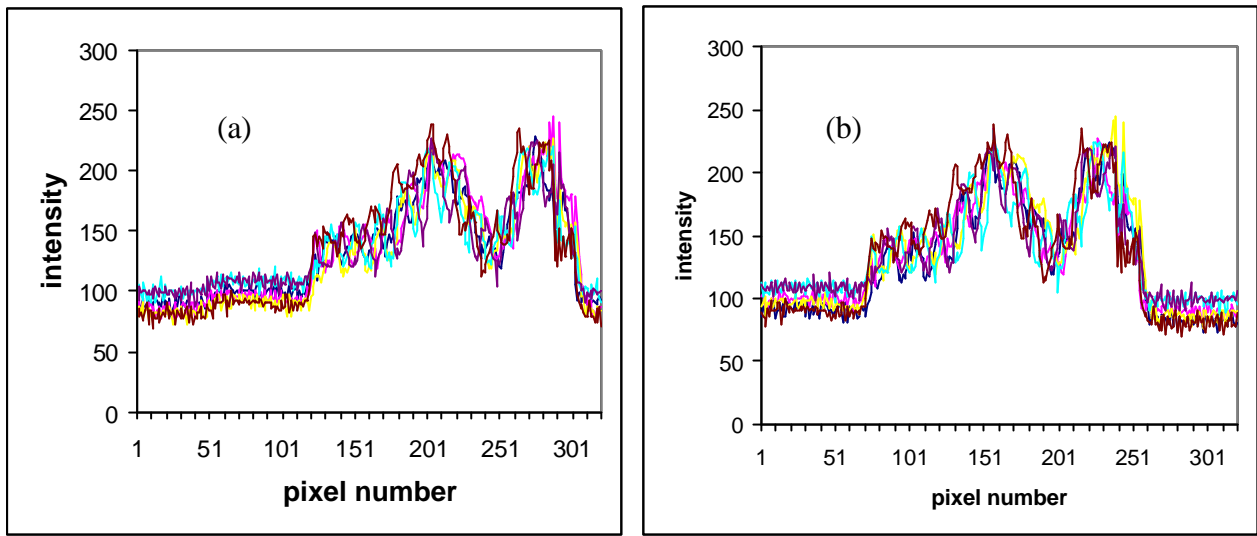


Figure 7 Fringe patterns of 20% ethanol at different temperatures (a)  $T_a = 21^\circ\text{C}$ , (b)  $T_a = 140^\circ\text{C}$

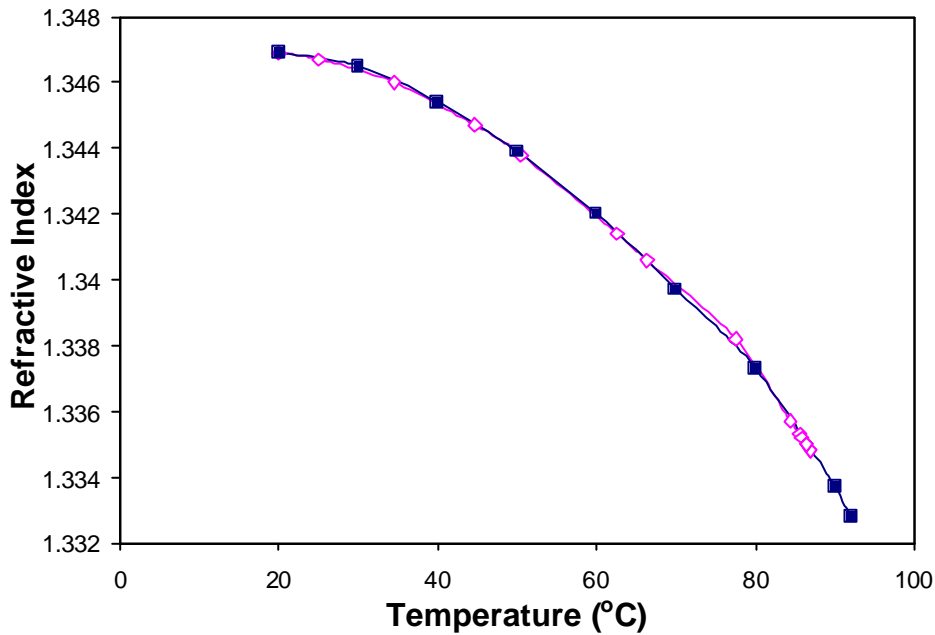


Figure 8 The calibration curve of 20% ethanol (80% water)

Pure water, 20% ethanol, 40% ethanol, 60% ethanol, 80% ethanol and pure ethanol were measured using as the experimental procedures described above. The results of measurements are shown in Figure 9 and Figure 10. Figure 9 shows that when the air temperature increased, the droplet diameters of all liquids decreased because of evaporation. Also the diameter of the pure ethanol droplet changed faster than the diameter of water droplet. With the increasing of the percent of ethanol in the mixture, the changing rate of the droplet was faster. This is also because that the ethanol has higher volatility but lower boiling temperature than water. Therefore, it is easy to be evaporated.

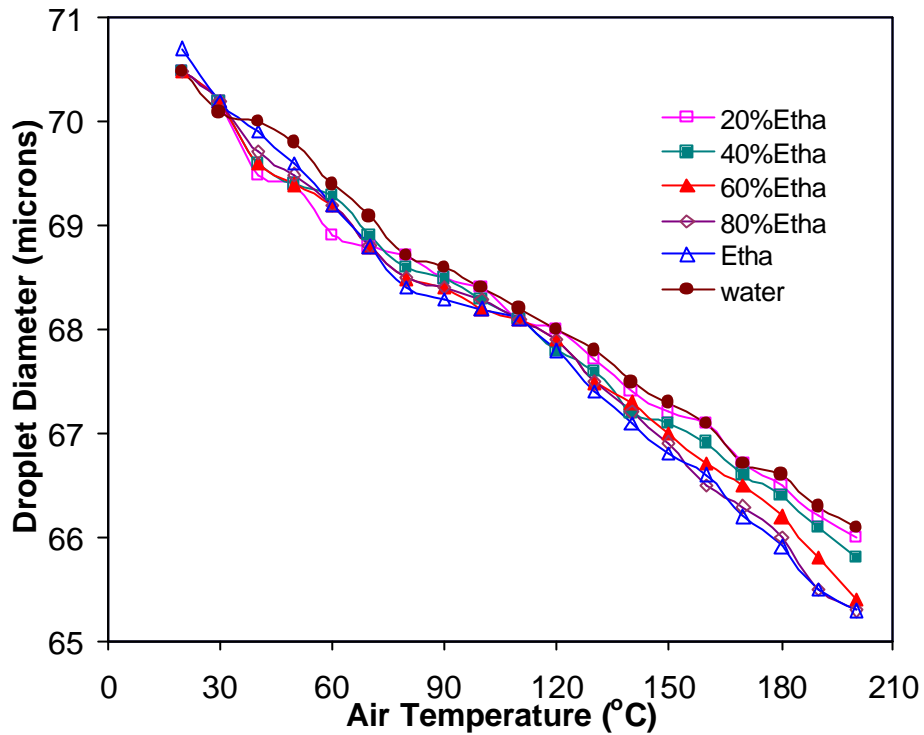


Figure 9 The measurement results of droplet diameter via different air temperatures

Figure 10 also shows that when field temperature was low, the temperature changing rate of pure ethanol was the fastest among all of the mixtures and pure water.

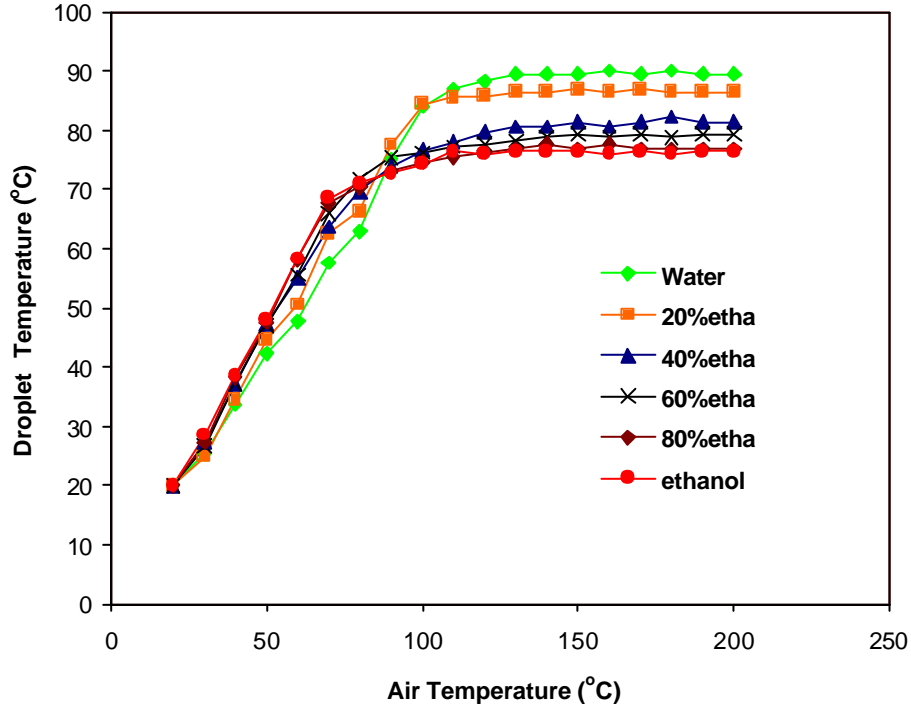


Figure 10 The measurement results of droplet temperatures via different air temperatures

With the increasing of the composition of ethanol in the multicomponent liquid droplet, the changing rate also increased. It was also found that when the air temperature field reached a critical level and above, such as around 150°C for water, the measured droplet temperatures of all components became unchanged. One might explain this phenomenon assuming

the droplets of liquids had already reached their saturated status uniformly within individual droplets when the air temperature reached the critical value. After that, the droplets were assumed to evaporate under homogenously saturated temperature condition. Under this assumption, the temperatures of droplets could not increase any more. However, it was also found that all of the droplet temperatures under this condition were below the boiling temperatures of the liquids according to the properties of liquids. Because the temperature measurement technique (rainbow refractometer) has been calibrated for all the liquids prior measurements started, it is reasonable to explain that the measured temperature differences were caused by the temperature gradient within the droplets. Therefore, the previous assumption regarding uniform temperature within the droplets becomes unjustifiable. As a result, it is reasonable to assume that the evaporating interface may act as mass sources and heat sinks while the liquid within the droplets may act as a heat source when its temperature reaches a certain stage. The balance between the heat sink effect of evaporating interface and the heat conduction from the outside of hot air causes the liquid temperature within the droplet reaches a constant temperature which is below the boiling temperature of the liquid.

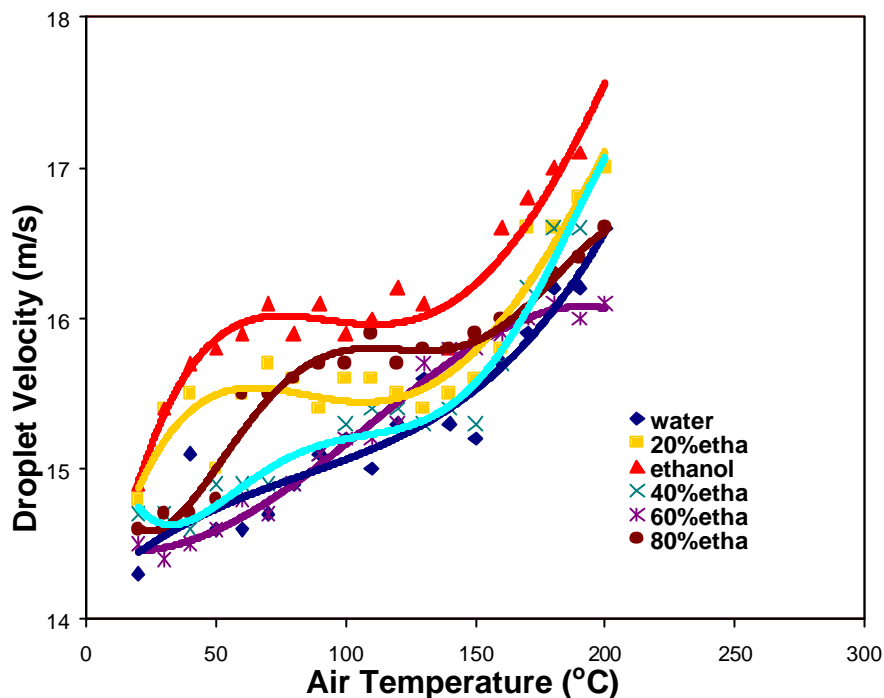


Figure 11 The measurement results of droplet temperatures via different air temperatures

Figure 11 shows the relationship between the air temperatures and the droplet velocities. Increasing the air temperature resulted in increasing the droplet velocity. This might be caused by the reducing the viscosity resistance by increasing the air temperature. A nonlinear effect for pure ethanol was also observed.

## CONCLUSIONS

Two novel optical techniques were used for investigating evaporating multicomponent microdroplets. With the newly developed optical diagnostic method, it is possible to probe the evaporation rate of multi-component droplets under various temperature conditions. The behaviors of single- and multi-component microdroplets were studied. It was found that the droplet temperature could never reach uniform within the droplet during the evaporating process unlike explained by previous researchers. This finding can be explained using the temperature gradient within the evaporating microdroplet which may be caused by the heat sink effect of the evaporating interface.

## ACKNOWLEDGEMENT

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