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IR-Thermography of Evaporating Acoustically Levitated Drops

R. Tuckermann,[1,2] S. Bauerecker,[3] and H. K. Cammenga[3]

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The surface temperature of evaporating acoustically levitated drops has been monitored by IR-thermography. The thermography system has been calibrated by simultaneous measurements with a thin thermocouple. The integral IR-emission coefficients of a few pure liquids have been derived. Evaporation processes of pure liquids and of binary mixtures of liquids have been studied with levitated drops. Using drop surface temperatures, we were able to calculated analytically binary gas-diffusion coefficients of liquids in ambient air.

KEY WORDS: acoustic levitation; binary gas-diffusion coefficients; integral emissivity; IR-thermography; pure and binary mixtures of liquids; surface temperature.

1. INTRODUCTION

Evaporation processes of single drops have been studied in many experimental, theoretical, and numerical investigations [1–3]. Besides the time evolution of the drop size, the surface temperature of the drop is one of the main parameters to characterize the evaporation process. The temperature of a suspended drop can be measured by different techniques, for example, with a thin thermocouple or by an infrared (IR)-thermography system. The latter offers the advantage of a contactless and non-intrusive method.

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[1] Institut für Küstenforschung/Umweltchemie, GKSS-Forschungszentrum Geesthacht GmbH, Max-Planck-Strasse 1, D-21502 Geesthacht, Germany.

[2] To whom correspondence should be addressed. E-mail: rudolf.tuckermann@gmx.de

[3] Institut für Physikalische und Theoretische Chemie, Technische Universität Braunschweig, Hans-Sommer-Strasse 10, D-38106, Braunschweig, Germany.

For a correct analysis of infrared images, the radiation transfer between the object and the detector has to be known, especially the integral emissivity,

$$\varepsilon = \int_{\lambda_1}^{\lambda_2} \varepsilon(\lambda, T)\, d\lambda = \int_{\lambda_1}^{\lambda_2} \frac{P_{\mathrm{real}}(\lambda, T)}{P_{\mathrm{black}}(\lambda, T)} d\lambda \approx \frac{P_{\mathrm{real}}(\Delta\lambda, T)}{P_{\mathrm{black}}(\Delta\lambda, T)} \tag{1}$$

of the emitting object within the spectral region, $\Delta\lambda = \lambda_2 - \lambda_1$, of the IR-thermography system. The emission coefficient $\varepsilon(\lambda, T)$ of an object as a function of temperature T and wavelength λ is the ratio of the radiance of a real body $P_{\mathrm{real}}(\lambda, T)$ to the radiance of a blackbody $P_{\mathrm{black}}(\lambda, T)$. From the radiance of a blackbody the temperature of the object can be derived, using Planck's law and taking into account the geometrical alignment of object and detector during the measurements.

Many IR-thermography systems are operating in a spectral range of 8–12 μm. The integral emissivities of various objects and materials in this spectral range are well documented in the literature [4], e.g., pure water at 20 °C has an integral emissivity of 0.96. But there is only a little information about the integral emissivities of other organic liquids [5–7]. Their values are generally assumed to be in the range between 0.9 and 1.0, close to the emissivity value of pure water [5].

In this investigation we have studied surface temperatures of acoustically suspended drops of various organic liquids and of water by an IR-thermography system in ambient air. Acoustic levitation is a useful and easy tool for suspending small liquid and solid samples in the pressure nodes of a stationary ultrasonic field. The sample diameters are typically in the range of approximately 0.1–5 mm, depending mainly on the frequency of the stationary ultrasonic field. An introduction to the technique of acoustic levitation with regard to its applications is given elsewhere [8, 9]. From our measurements of evaporating drops, qualitative and quantitative results have been obtained; for a few liquids the emission coefficients were determined. The surface temperatures of evaporating levitated drops of pure and mixed liquids have been measured, and binary gas-diffusion coefficients have been calculated. Processes of condensation, of two-phase separation, and of ice formation have been observed during drop evaporation.

2. EXPERIMENTAL

The experimental setup is shown in Figs. 1 and 2. Drops of various liquids with diameters in the range of 1–2 mm were suspended in the pressure nodes of a stationary ultrasonic field in an acoustic levitator. The drop temperature has been monitored by an IR-thermography system and

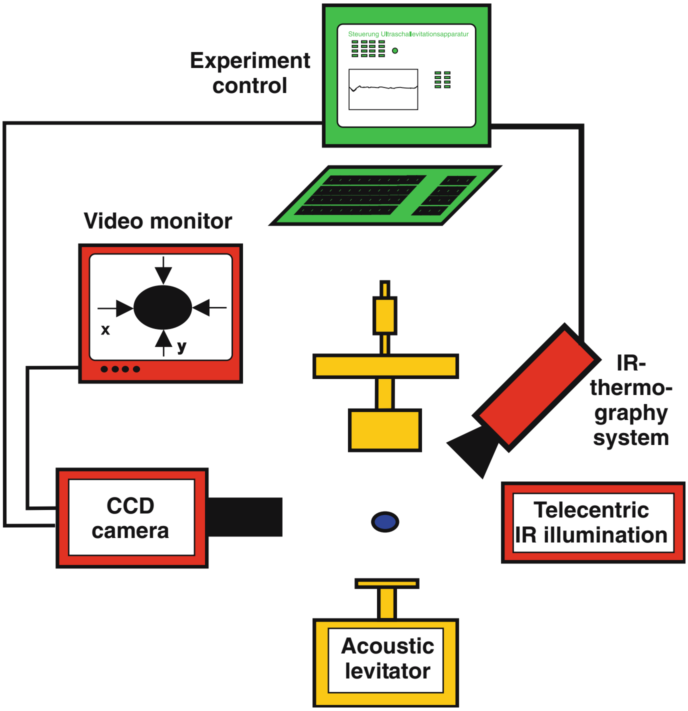

Fig. 1. Schematic diagram of the experimental setup.

a 0.25 mm diameter Ni–CrNi mantle thermocouple (Thermocoax GmbH, Hamburg, Germany). The drop size has been measured by an intelligent CCD camera from backlit images of the drop. The CCD-camera system is described in Ref. 10. The experiments were performed in ambient air at ambient temperature and pressure conditions. The temperature, pressure, and relative humidity (RH) of the gaseous environment were also measured during the experiments.

2.1. Acoustic Levitator

The one-axial acoustic levitator used in this investigation operates at 58 kHz corresponding to a wavelength of $\lambda = 5.7$ mm in ambient air. The stationary ultrasonic field is generated between an ultrasonic radiator and a concave reflector, positioned concentrically to the radiator at a distance

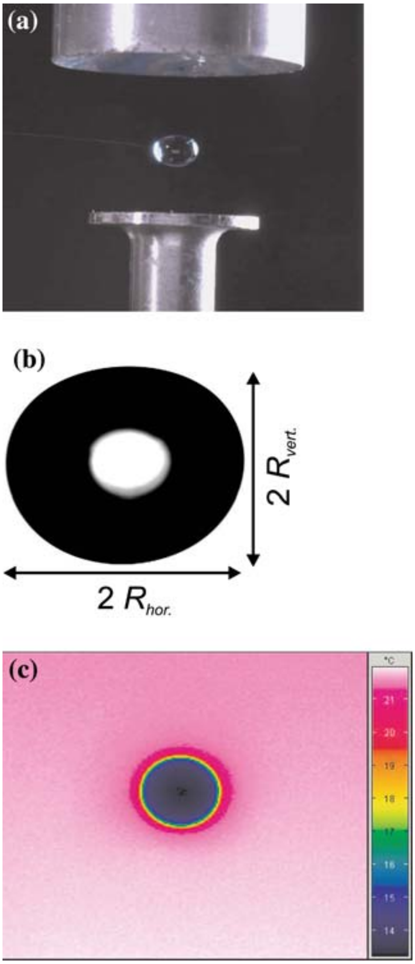

Fig. 2. (a) Photograph, (b) backlit image, and (c) IR image of an acoustically levitated water drop in ambient air (20 °C and 50% RH). The photo shows the drop in the stationary ultrasonic field of a 58-kHz acoustic levitator with the ultrasonic transducer at the bottom and the reflector at the top of the picture. From the backlit image the drop size is measured by an intelligent CCD-camera, and from the IR image the drop temperature can be derived.

of

$$d = n\frac{\lambda}{2}(\text{with } n = 1, 2, 3, \ldots) \tag{2}$$

in order to meet resonance conditions. Acoustic levitation is possible in each pressure node of the stationary ultrasonic field. In the present investigation the central pressure node was used for levitation at a distance between the radiator and reflector of $n = 5$. The sound pressure level inside the pressure nodes of the ultrasonic field has been determined to be approximately 160 dB [11, 12].

2.2. IR-Thermography System

The IR-thermography system (Model VarioScan 3011 of InfraTec GmbH, Dresden, Germany) operates in the wavelength range of 8–12 μm and is composed of an infrared camera, an MCT-detector cooled by liquid nitrogen, and a macro-lens. The latter allows a maximum space resolution of approximately 25 μm per pixel. The image frequency of the system is approximately 1 Hz, and the possible temperatures range from −30 to 1200 °C with a feasible temperature resolution of 0.1 °C. The whole system is operated by special software. For analyzing IR images, the software IribisPlus® (InfraTec GmbH, Dresden, Germany) was used.

2.3. Chemicals

The following chemicals were purchased from Merck KGaA, Darmstadt, Germany: methanol p.a., ethanol p.a., 1-butanol p.a., *n*-pentane p.a., and *n*-hexane p.a., formic acid, acetic acid, acetone, diethyl ether, chloroform, dichloromethane, pyridine, and from Fluka Feinchemikalien GmbH, Buchs, Switzerland: 1-propanol (purity ≥ 99% v/v), 1-pentanol (purity ≥ 99% v/v), 1-hexanol (purity ≥ 99% v/v), *n*-heptane (purity ≥ 99% v/v), *n*-octane (purity ≥ 99% v/v), *n*-nonane (purity ≥ 99% v/v), and *n*-decane (purity ≥ 99% v/v).

3. RESULTS AND DISCUSSION

3.1. Determination of Integral Emissivities

The temperatures derived from IR images of acoustically levitated evaporating drops using different values of the integral emissivity have been compared with the drop temperatures, which has been simultaneously measured by a thin thermocouple (see Fig. 3). The integral emissivities

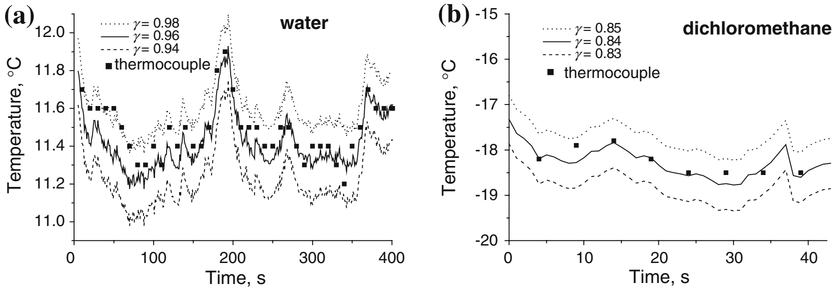

Fig. 3. Simultaneous temperature measurements of (a) an evaporating water drop and (b) a dichloromethane drop in ambient air (20 °C and 26% RH) with a thermocouple (squares) and with an IR-thermography system using different emissivity values ε for analysis of the IR-pictures. The best fits were obtained from $\varepsilon = 0.96$ for water and $\varepsilon = 0.84$ for dichloromethane (see also Table I).

leading to the best temperature fits are listed in Table I. In our analysis we have assumed that the surface temperature of an acoustically levitated drop measured by the IR-thermography system corresponds to its mean temperature measured by the thermocouple. This assumption is justified by the small size and the internal convection of acoustically levitated drops [2] which lead to a rapid temperature equilibration. Further, there are heat inputs from the stationary ultrasonic field and the thermocouple into the levitated drop which cause an additional warming of the drop in the range of a few tenths to 1 °C.

The determined value of the integral emissivity of water ($\varepsilon = 0.96$) is in good agreement with literature data, while the values obtained for the organic solvents are a little smaller than reference values: for *n*-decane [5]

Table I. Evaluated Emissivity Values of Different Organic Solvents in the Spectral Range of 8–12 μm. (The uncertainty of the integral emissivity is about ±0.01.)

Solvent	Integral emissivity
water	0.96
ethanol	0.92
1-propanol	0.94
n-pentane	0.84
n-heptane	0.87
n-octane	0.86
dichloromethane	0.84
diethyl ether	0.90

and 1-propanol [6] a value of 0.95 is confirmed in the literature, while we obtained 0.84–0.87 for *n*-heptane, *n*-octane, and *n*-nonane and 0.94 for 1-propanol. In general, ε-values for organic liquids are assumed to fall in the range of 0.9–1.0 [5]. For further analysis of those IR images for which we have done no additional temperature measurements by thermocouples, we have taken an ε-value of 0.95. This may cause a systematic maximum deviation of approximately 1 °C in the absolute temperature determination. This error has to be taken into account for further quantitative analysis.

3.2. Surface Temperature of Evaporating Drops and Determination of Binary Gas-Diffusion Coefficients

Evaporation processes of suspended liquid drops in a gaseous environment can be described by a linear decrease of the drop surface area S with time t [13];

$$S = S_0 - K\,t, \tag{3}$$

where the proportionality factor K is a function of the liquid density ρ_l, the molecular mass M, the binary gas-diffusion coefficient D_ab of the vapor in the surrounding gas, the partial vapor pressure p, and the temperature T at the drop surface (subscript s) and in the gaseous environment (subscript ∞):

$$K = \frac{8\pi D_\mathrm{ab} M}{\rho_\mathrm{l} R}\left(\frac{p_\mathrm{s}}{T_\mathrm{s}} - \frac{p_\infty}{T_\infty}\right)\frac{Sh}{2}. \tag{4}$$

where R is the universal gas constant and Sh is the Sherwood number, describing the ratio of mass transfer with and without convection in the gas. Within the stationary ultrasonic field of the acoustic levitator used in this investigation, acoustic streaming leads to a higher mass transfer [14]. For our experimental conditions we have worked with $Sh = 3.5$ [7].

Examples of surface area and temperature measurements of evaporating acoustically levitated drops of pure organic liquids and water in ambient air are given in Fig. 4. Corresponding data of the homologues of *n*-alkanes and 1-alkanols have been published in Ref. 7. The values of surface temperature T_s and evaporation constant K have been determined at the beginning of the evaporation process and have been summarized in Table II. Using Eq. (4), from these data the binary gas-diffusion coefficients D_ab of the vapors of the organic solvents in ambient air or gas can be analytically calculated. The results have also been listed in Table II.

Considering the uncertainties of the measurements and the analysis, the results are in agreement with calculated data from the literature [15].

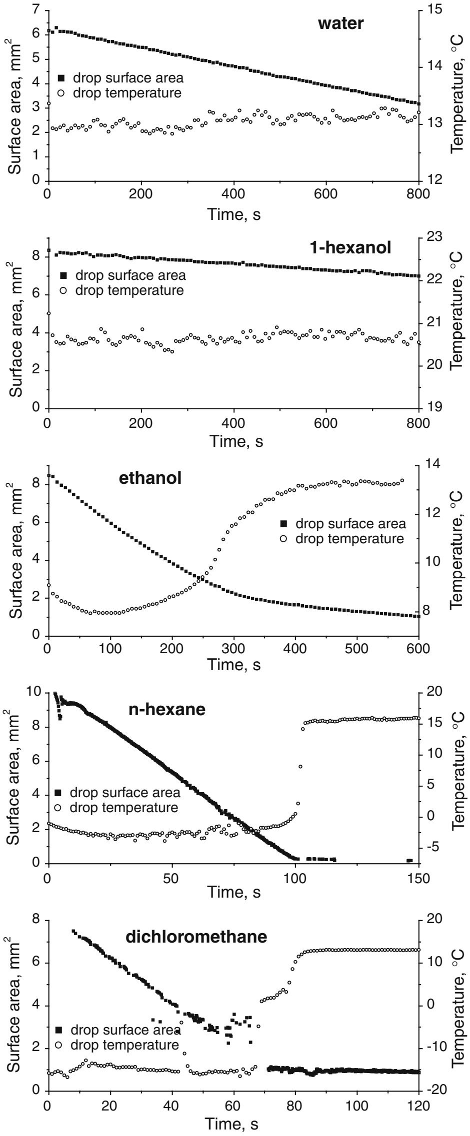

Fig. 4. Time evolution of surface area and temperature of acoustically levitated drops, evaporated in ambient air at 21 °C and 35–40% RH.

Table II. Surface Temperatures T_s and Evaporation Constant K of Acoustically Levitated Drops of Various Pure Organic Solvents at the Beginning of Their Evaporation in Ambient Air ($T_\infty = 21\,^\circ\mathrm{C}$ and 35–40% RH). (The uncertainty of the surface temperature is about 0.5 °C. The experimental binary gas-diffusion coefficients $D_{ab}^{\mathrm{Exp.}}$ have been calculated by using Eqs. (3) and (4), whereas the $D_{ab}^{\mathrm{Lit.}}$ values have been derived from the literature [15].)

Solvent	Surface temperature T_s(°C)	Evaporation constant K (mm^2·s^{-1})	Binary gas-diffusion coefficient of the solvent vapor in ambient air	
			$D_{ab}^{\mathrm{Exp.}}$ (cm^2·s^{-1})	$D_{ab}^{\mathrm{Lit.}}$ (cm^2·s^{-1})
n-pentane	−18.8	0.20 ± 0.01	0.084 ± 0.006	0.082
n-hexane	−3.8	0.106 ± 0.005	0.079 ± 0.004	0.075
n-heptane	8.3	0.049 ± 0.002	0.071 ± 0.003	0.069
n-octane	15.8	0.020 ± 0.001	0.062 ± 0.003	0.064
n-nonane	19.6	0.008 ± 0.001	0.062 ± 0.008	0.060
methanol	−0.5	0.045 ± 0.005	0.147 ± 0.015	0.157
ethanol	7.9	0.032 ± 0.003	0.106 ± 0.010	0.120
1-propanol	11.7	0.014 ± 0.002	0.101 ± 0.014	0.100
1-butanol	17.2	0.007 ± 0.001	0.095 ± 0.013	0.087
1-pentanol	20.4	0.004 ± 0.001	0.091 ± 0.020	0.079
1-hexanol	20.7	0.0015 ± 0.0003	0.087 ± 0.018	0.072
acetone	−8.4	0.085 ± 0.005	0.097 ± 0.006	0.103
diethyl ether	−19.5	0.16 ± 0.01	0.084 ± 0.005	0.087
formic acid	13.6	0.036 ± 0.010	0.16 ± 0.04	0.140
acetic acid	8.0	0.020 ± 0.005	0.14 ± 0.07	0.110
chloroform	−5.4	0.082 ± 0.003	0.087 ± 0.003	0.089
dichloromethane	−15.8	0.097 ± 0.005	0.087 ± 0.005	0.102
pyridine	19.0	0.023 ± 0.005	0.080 ± 0.017	0.093

The accuracy of the analysis may be improved by taking into account the effect of the stationary ultrasonic field on the evaporation process of acoustically levitated drops more precisely. Therefore, from a numerical analysis of the drop shape the sound pressure level of the acoustic field and the values of the acoustic streaming as well as the Sherwood number can be determined with higher accuracy [2].

Nevertheless, the main effort of the present investigation is that we have measured the drop temperature directly and contactless via IR-thermography, while in most of the other investigations this parameter has been numerically calculated. From the temperature measurements we were able to derive the binary gas-diffusion coefficients analytically by using Eqs. (3) and (4).

Some more qualitative aspects can be deduced from the results given in Fig. 4: a linear decline in drop surface area with a constant drop temperature during the whole experiment as predicted from Eq. (3) can only be seen in the case of water and 1-hexanol in Fig. 4. In the other cases some humidity from the ambient air has condensed on the surface of the evaporating drops, influencing the time evolution of the drop surface area and temperature; as a result of the good solubility of water in ethanol and the higher volatility of ethanol, a drop of initially pure ethanol slowly develops to a pure water drop during the evaporation process in ambient air. For the cases of *n*-hexane and dichloromethane, condensed water has formed a separate phase inside the evaporating drops of *n*-hexane and dichloromethane. Therefore, a small water drop has remained, after all of the organic solvent had been evaporated, indicated by a jump in the evolution of the drop temperature T_s and the evaporation constant K. For the case of dichloromethane, where the drop temperatures have decreased far below 0 °C, the condensed water has formed an ice crystal inside the dichloromethane drop. After all dichloromethane had been evaporated, the ice crystal has melted and a small drop of liquid water finally remained.

3.3. Surface Temperature of Evaporating Drops of Binary Liquid Mixtures

Depending on their mixing ratio, binary mixtures of liquids can be totally or partially miscible or totally immiscible. An immiscible mixture of two liquids separates into two phases; in an acoustically levitated drop, the higher-density phase is mostly covered by the low-density phase. In Fig. 4 two examples of an immiscible mixture have already been given; water condensing on the drop surface from ambient air has formed a separate phase inside an *n*-hexane or dichloromethane drop. The temperature of those drops of immiscible liquids is given by the evaporation temperature of the outer compound with a lower density, i.e., *n*-hexane and dichloromethane in our example.

In the case of miscible liquids, the drop temperature is changing with the mixing ratio of the two liquids during the evaporation process. This has been clearly demonstrated in Fig. 4 by the example of an evaporating drop of ethanol in ambient air. The mixing ratio has continuously changed, when water was condensing on the drop surface while ethanol was evaporating from the drop. Starting with a pure ethanol drop with a characteristic temperature of approximately 8 °C, the remaining drop at the end of the experiment had a temperature of approximately 13 °C, corresponding to the evaporation temperature of an acoustically levitated pure water drop in ambient air of 20 °C and 40% RH.

The evaporation processes of drops containing miscible liquids can obviously be described no longer by Eq. (3). A quantitative analysis of the evaporation of acoustically levitated drops of binary mixed liquids is given in Ref. 3.

For binary liquid mixtures of water/methanol, water/1-propanol, and pyridine/formic acid, we have measured the evaporation temperature as a function of their mixing rates. The results are presented in Fig. 5. In addition, the composition-dependent boiling temperatures of these mixtures [16] have also been plotted in Fig. 5. The water/methanol system is an example of an ideal mixture, while water/1-propanol and pyridine/formic acid are azeotropic mixtures with a minimum and maximum, respectively, in the boiling and evaporation temperatures. The qualitative correspondence of the boiling temperature and the drop evaporation temperature as a function of the mixing ratio in all three systems is remarkable.

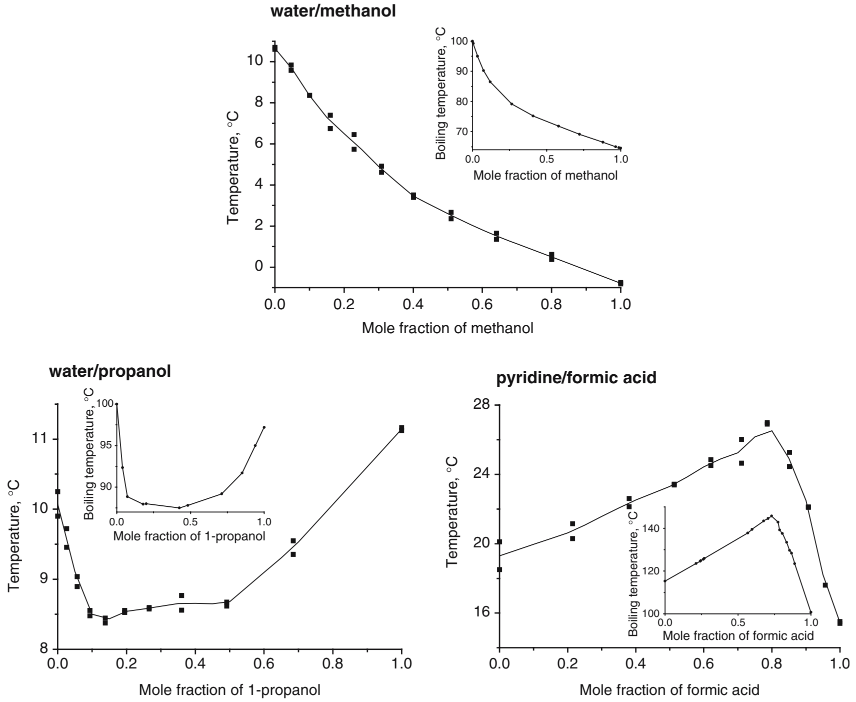

Fig. 5. Temperatures of evaporating drops of binary mixed solvents in ambient air as a function of their composition. The relative humidity of the ambient air was 29% in the case of evaporating water/methanol drops, 25% in the case of water/1-propanol drops, and 45% in the case of pyridine/formic acid drops. The qualitative correspondence of the boiling temperatures of the mixtures (small figures) compared to the drop evaporation temperature is remarkable.

4. CONCLUSION

The temperature of an evaporating and levitated drop is an important parameter for characterizing the evaporation process. In most of the previous studies related to the evaporation of suspended drops, the drop temperature had been numerically calculated. In the present investigation we have demonstrated the feasibilities of the contactless measurement of the drop temperature via IR-thermography. For calibration of the system the integral emissivity must be known from the literature or must be determined experimentally. The evaporation processes of acoustically levitated drops of pure liquids and binary liquid mixtures have been characterized by the drop temperature. For 18 solvents, the binary vapor/air-diffusion coefficients have been derived from the drop temperature, and the results obtained are in accordance with data from the literature.

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