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Multi-component droplet growth. I. Experiments with supersaturated *n*-nonane vapor and water vapor in methane

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Droplet growth rates of droplets suspended in methane gas and supersaturated water and/or *n*-nonane vapor are experimentally determined. The experiments are performed by applying the nucleation pulse principle using a modified shock tube. The droplets are optically detected using a combination of constant angle Mie scattering and light extinction measurements. From the analysis of the droplet growth rates in the binary systems the diffusion coefficients of water in methane and *n*-nonane in methane have been determined at two different conditions, being 11 bar and 242 K, and 44 bar and 247 K. The droplet growth rates in the ternary system have been determined at the same two conditions. From the analysis of these experiments it is evident that supersaturated water vapor does not condense onto *n*-nonane droplets while supersaturated *n*-nonane vapor does condense onto water droplets. This can be related to the wetting properties of liquid water on liquid *n*-nonane and vice versa. © 2004 American Institute of Physics. [DOI: 10.1063/1.1751191]

I. INTRODUCTION

The growth of multi-component droplets is encountered both in nature and many industrial applications. In nature, clouds of water droplets can be formed which contain different kinds of acids. Similarly, this can occur in the waste outlets of industrial plants. At very different conditions, liquid droplets can be formed in pipelines containing mixtures of gas and vapor. An example of such a mixture of gas and vapor is natural gas. Often, one aims to prevent the formation of droplets in pipelines, because these can damage, for instance, valves. In other cases, the formation of droplets is desirable, like in the newly developed supersonic gas separator for the natural gas industry.¹ In this apparatus, liquid droplets are formed at relatively high methane pressures (tens of bars), low temperatures (approximately 200 to 250 K), and large supersaturation of many different vapor components. These vapor components can roughly be divided into the *a*-polar hydrocarbon chains and the polar water. In order to better understand the growth processes of the liquid droplets at these conditions a research program was started at the Eindhoven University of Technology.^{2,3} Using a modified shock tube, the growth of liquid droplets suspended in supersaturated mixtures of methane, *n*-nonane, and/or water is systematically investigated at the conditions of interest. In a series of two consecutive papers, we will report on the analysis of the experimental results, and on the theoretical treatment of multi-component layered droplet growth.⁴

II. EXPERIMENTS

The experiments are performed by applying the nucleation pulse principle using a modified shock tube.^{5,6} The setup is schematically shown in Fig. 1. The gas–vapor mixture in the high pressure section (HPS) is prepared using the dedicated mixture preparation device (MPD) which has been

described in detail in a previous paper.² The resulting pressure history at the end wall of the HPS when the diaphragm between the HPS and the low pressure section (LPS) of the shock tube has been ruptured is shown in Fig. 2. The pressure at the end wall of the HPS is recorded using a Kistler 603B dynamic pressure transducer. The temperature (not shown) is calculated from the pressure history using isentropic adiabatic relations.⁷ Also shown in Fig. 2 is the resulting supersaturation *S* of the vapor in the HPS. The supersaturation is defined as

$$S = \frac{y}{y^{eq}}, \quad (1)$$

where y^{eq} is the equilibrium molar vapor fraction at the given conditions. The small pressure dip after the large expansion (see Fig. 2) is formed due to wave reflections at the local widening in the LPS, just behind the diaphragm (Fig. 1). A detailed description of these wave reflections and the resulting pressure history at the end wall of the HPS is given by Looijmans *et al.*⁶ The molar vapor fraction in the HPS is constant and given by the initial conditions. The change in the supersaturation *S* is therefore entirely due to the change of the equilibrium vapor fraction y^{eq} . During the pressure dip the equilibrium molar vapor fraction is at its lowest value, resulting in the peak in the supersaturation. During this state of high supersaturation the homogeneous nucleation rate (i.e., formation of new stable droplets) is orders of magnitude larger than after the pulse (typically 10^9 to 10^{13} stable nuclei per cubic meter are formed during the pulse). The amount of new droplets formed after the pulse of high supersaturation is negligible. However, the system remains in a supersaturated state after the pulse, resulting in the growth of the stable nuclei formed in the pulse. The nucleation pulse

FIG. 1. Schematic view of the experimental setup.

method effectively decouples the nucleation and growth stage of the droplets, resulting in a (nearly) monodispersed cloud of growing droplets.

The size and number density of the droplets are determined using a combination of 90° light scattering and light extinction measurements from a laser beam of wavelength $\lambda = 514.2$ nm. The ratio of the droplet number density and the duration of the nucleation pulse gives the steady nucleation rate for each experiment. These experimentally obtained nucleation rates were the subject of a previous paper.² From the light scattering signals of these experiments, recorded by the photomultiplier (see Fig. 1), the droplet growth curves can be determined. In Fig. 3 a typical scattering signal of *n*-nonane droplets in methane gas at 11 bar and 242 K is shown. The scattering intensity varies in time as the droplets simultaneously grow larger. Using Mie theory, the scattering intensity can be calculated as a function of the size of the droplets.^{8–10} The result is shown in Fig. 4, where α is a dimensionless size parameter defined as

$$\alpha = \frac{2\pi r_d}{\lambda}, \quad (2)$$

where r_d is the radius of the droplets. Consequently, by relating each extremum in the experimentally obtained time dependent plot to the same extremum in the theoretical size-dependent plot a droplet growth curve can be constructed.¹¹

FIG. 2. Pressure and supersaturation history at the end wall of the HPS.

FIG. 3. Experimentally observed scattering intensity of *n*-nonane droplets in methane.

III. DROPLET GROWTH

From the analysis of the nucleation results we have learned that water and *n*-nonane nucleate independently, which is also confirmed by the results obtained by Wagner and Streyl.^{2,12} We will now proceed to analyze the growth results of these experiments. First, we will analyze the growth rates of droplets in systems of either supersaturated water vapor or supersaturated *n*-nonane vapor in methane. We will then compare these droplet growth rates in the binary systems to the droplet growth rates in ternary systems where both water and *n*-nonane are supersaturated in methane.

A. Binary mixtures

As discussed in Sec. II, droplet growth curves have been obtained from the light scattering signals, for each individual experiment. In Fig. 5 the growth curve resulting from the scattering signal shown in Fig. 3 is plotted. The nucleation pulse is positioned at about 7 ms, after which the droplets start to grow. The smallest detectable droplet radius corresponds to the maximum of the first Mie peak: about $0.16 \mu\text{m}$, for both *n*-nonane and water droplets. Because of the high carrier gas pressure (>10 bar), the Knudsen number

FIG. 4. The scattering intensity of *n*-nonane droplets in methane for the geometry of our setup.

FIG. 5. The droplet radius squared as a function of time, for a typical experiment of supersaturated *n*-nonane in methane, at 11 bar and 242 K.

($\text{Kn} = d/r_d$, where d is the mean free path of the molecules) is small at this condition ($\text{Kn} < 0.015$), and the growth of the droplets is diffusion controlled. For diffusion controlled growth the increase of the droplet surface is proportional to time, as can be seen in Fig. 5. In the case of steady growth the factor of proportionality can be obtained by equating the diffusion controlled molar flux $\dot{M}$ expressed as⁴

$$\dot{M} = -4\pi r_d \rho^G D (y - y_{r_d}^{eq}), \quad (3)$$

to the molar flux obtained from the time-dependent increase of the droplet

$$\dot{M} = \frac{d}{dt} \left[\frac{4}{3} \pi r_d^3 \rho^L \right]. \quad (4)$$

In these equations ρ^G is the molar gas phase density, ρ^L is the molar liquid density of the vapor, $y_{r_d}^{eq}$ is the equilibrium vapor fraction at the surface of the droplet, and D is the diffusion coefficient of the vapor through the gas. By equating the fluxes one obtains

$$\frac{dr_d^2}{dt} = \frac{2\rho^G D}{\rho^L} (y - y_{r_d}^{eq}). \quad (5)$$

FIG. 6. Surface growth rate of water in methane at 11 bar and 242 K as a function of the molar water fraction. The point in the abscissa corresponds to the calculated phase equilibrium at the given pressure and temperature, using the CPA equation of state.

FIG. 7. Surface growth rate of water in methane at 44 bar and 247 K as a function of the molar water fraction. The point in the abscissa corresponds to the calculated phase equilibrium at the given pressure and temperature, using the CPA equation of state.

At constant temperature and pressure, as long as there is no significant depletion, the right-hand side of this equation is constant. Throughout the rest of this paper we will refer to dr_d^2/dt as the surface growth rate, omitting a factor 4π . The surface growth rate is proportional to the vapor fraction, and the constant of proportionality is given by $2\rho^G D/(\rho^L)$. In Figs. 6–9 the surface growth rates of *n*-nonane in methane and water in methane are shown as functions of the vapor fraction, at the two different conditions studied. The surface growth rate equals zero when the vapor fraction equals the equilibrium vapor fraction, at the given temperature and pressure. The temperature and pressure are obtained by averaging over the growth stage of all the relevant experiments. From this averaged temperature and pressure an average equilibrium fraction is calculated using the CPA eos (cubic plus association equation of state), for each condition considered.^{13–15} Then, a line is drawn from this equilibrium fraction through the experimental data. Within the accuracy of the experiments, this is possible for all experimental conditions investigated. This provides an independent check of

FIG. 8. Surface growth rate of *n*-nonane in methane at 11 bar and 242 K as a function of the molar *n*-nonane fraction. The point in the abscissa corresponds to the calculated phase equilibrium at the given pressure and temperature, using the CPA equation of state.

FIG. 9. Surface growth rate of n -nonane in methane at 44 bar and 247 K as a function of the molar n -nonane fraction. The point in the abscissa corresponds to the calculated phase equilibrium at the given pressure and temperature, using the CPA equation of state.

the calculated vapor fractions, and therefore of the quantitative reliability of the CPA eos, which we will use in our calculations. Especially at 44 bar and 247 K for n -nonane in methane the agreement is very good. At the other conditions the supersaturation is higher, making the determination of the equilibrium vapor fraction from the experimental data less accurate, due to the necessary extended range of extrapolation. It is noteworthy that the setup provides a means to experimentally determine equilibrium vapor fractions. By varying the depth of the nucleation pulse (by varying the depth of the widening), the growth of the droplets can be determined at larger and smaller supersaturation (i.e., vapor fraction). Hence, the cutoff in the abscissa in Figs. 6–9 can be determined with higher accuracy, resulting in accurate experimentally determined equilibrium vapor fractions.

The diffusion coefficient can be determined experimentally, since it is directly related to the slopes of the lines drawn in Figs. 6–9. The experimentally obtained diffusion coefficients are listed in Table I for each binary mixture and each condition. Also shown in this table are values for the diffusion coefficient when using the semi-empirical Fuller correlation.¹⁶ The values obtained from the Fuller correlation differ less than 15% from the experimentally obtained values, which is in agreement with the given accuracy of the Fuller correlation.

TABLE I. Experimentally determined diffusion coefficients of n -nonane in methane and water in methane.

	$n\text{-C}_9\text{H}_{20}\text{-CH}_4$		$\text{H}_2\text{O-CH}_4$	
	$D_{\text{exp}} (\text{m}^2 \text{s}^{-1})$	$D_{\text{Full}} (\text{m}^2 \text{s}^{-1})$	$D_{\text{exp}} (\text{m}^2 \text{s}^{-1})$	$D_{\text{Full}} (\text{m}^2 \text{s}^{-1})$
11 bar, 242 K	38.32E-8	45.03E-8	13.88E-7	15.76E-7
44 bar, 247 K	10.17E-8	10.46E-8	31.52E-8	36.42E-8

FIG. 10. Surface growth rate of droplets in mixtures of supersaturated n -nonane and water in methane as a function of the molar water fraction, at 11 bar and 242 K. Also shown is the surface growth rate of the binary water/methane mixture.

B. Ternary mixtures

For the experiments with the ternary mixtures of supersaturated n -nonane and water in methane similar growth curves are obtained as for the binary experiments. The ternary experiments were conducted such that the molar fraction of one supersaturated vapor component was kept approximately constant, while the molar fraction of the other component was increased stepwise.

We will first consider the case in which the supersaturation (i.e., molar vapor fraction) of n -nonane is kept constant, while the molar fraction of water is increased. In Figs. 10 and 11 the surface growth rate of the droplets in these mixtures is shown as a function of the molar fraction of water, at 11 bar and 242 K and 44 bar and 247 K, respectively. As shown in an earlier paper, n -nonane and water nucleate independently.^{2,12} Therefore, when n -nonane is the dominating nucleating vapor (i.e., it has the highest nucleation rate at the given conditions), there are orders of magnitude more n -nonane droplets than water droplets at the beginning of the growth stage. From Figs. 10 and 11 one can then conclude

FIG. 11. Surface growth rate of droplets in mixtures of supersaturated n -nonane and water in methane as a function of the molar water fraction, at 44 bar and 247 K. Also shown is the surface growth rate of the binary water/methane mixture.

FIG. 12. Surface growth rate of droplets in mixtures of supersaturated n -nonane and water in methane as a function of the molar n -nonane fraction, at 11 bar and 242 K. Also shown is the surface growth rate of the binary n -nonane/methane mixture.

that the supersaturated water vapor does not grow onto the n -nonane droplets, since the droplet growth rate is independent of the molar fraction of water. Similar behavior has also been observed by Rudolf and Wagner.¹⁷

Next, we will consider the growth rates in supersaturated mixtures of n -nonane and water when the supersaturation of water is kept fixed. Hence, water is the dominant nucleating vapor, and the number of water droplets at the beginning of the growth stage is orders of magnitude larger than the number of n -nonane droplets. In Figs. 12 and 13 the surface growth rates of the droplets are now shown as functions of the molar n -nonane fraction, at 11 bar and 242 K and 44 bar and 247 K, respectively. These figures show a remarkable behavior. The droplet growth rates in the mixtures of supersaturated n -nonane and water in methane increase when the molar fraction of n -nonane is increased beyond a minimum value. As the molar vapor fraction of n -nonane increases, the droplet growth rate in the ternary mixture approaches the droplet growth rate of the supersaturated binary n -nonane/methane mixture.

FIG. 13. Surface growth rate of droplets in mixtures of supersaturated n -nonane and water in methane as a function of the molar n -nonane fraction, at 44 bar and 247 K. Also shown is the surface growth rate of the binary n -nonane/methane mixture.

FIG. 14. Cross-section of a newly condensed phase B onto a condensed phase C , with contact angles α , β , and γ .

IV. SIMULTANEOUS GROWTH

We will now try to explain the origin of this different growth behavior. In the case that both n -nonane and water are supersaturated in the methane carrier gas, different nucleation processes can occur. First, n -nonane and water could form a single nucleus together, since both components would rather be in the liquid phase. However, n -nonane and water are very poorly miscible and, as was shown by Peeters *et al.*, and Wagner and Strey, co-nucleation will not occur.^{2,12} Besides homogeneous nucleation, heterogeneous nucleation could be a possible route for the nucleation process. Heterogeneous nucleation involves the formation of a cluster on a substrate, in contrast to homogeneous nucleation, which involves the formation of a cluster in the parent phase. The process of heterogeneous nucleation can be described in a similar way as homogeneous nucleation, by determining the energy difference between two states.¹⁸ The first state only contains the vapor and the substrate, while in the second state, part of the vapor molecules have formed a cluster on the substrate.

A. Contact angles

The wetting properties and formation of contact angles of liquid drops on substrate is far from trivial and an extensive literature is available dealing with this subject (see, for instance, Ref. 19). In order to gain more insight into the current problem at hand we will use a relatively simple approximation. In Fig. 14 a cross section is shown of a newly condensed phase B which has heterogeneously nucleated onto substrate C . The form of the lens on the substrate depends on the ratios of the different interfacial tensions. The angles α , β , and γ along the line of three phase contact are determined by the ratios of the interfacial tensions. Using Neumann's triangle, which can be obtained by setting the net force on the three-phase line equal to zero, the angles between the phases can be obtained.²⁰ For instance, the angle β is given by

$$\cos \beta = \frac{1}{2} \left(\frac{(\sigma^{AC})^2}{\sigma^{BC} \sigma^{AB}} - \frac{\sigma^{BC}}{\sigma^{AB}} - \frac{\sigma^{AB}}{\sigma^{BC}} \right). \quad (6)$$

The angles α and γ are obtained using similar expressions. In our case, the interfacial tension between the liquid n -nonane and water is approximated using Antonow's rule

FIG. 15. Wetting angles of n -nonane on water (β_n) and water on n -nonane (β_w).

$$\sigma_{nw} = |\sigma_w - \sigma_n|, \quad (7)$$

where σ_w and σ_n are the pure component surface tensions.²⁰ Now, the contact angle β can be calculated for the case that the lens consists of water and the substrate consists of n -nonane, and vice versa. In all cases, the gaseous phase mainly consists of methane. In Fig. 15 the angle β is shown as function of the temperature, for the two different methane pressures. Above approximately 257 K n -nonane is completely wetting on water and water is completely nonwetting on n -nonane. At the growth conditions studied here, being 242 K and 11 bar, and 247 K and 44 bar, the calculated contact angle of n -nonane on water is about 30°. The actual wetting angles will probably be smaller than the ones that are calculated here, because of the approximation that was made for the interfacial tension between liquid n -nonane and water. In the real system the presence of methane will most probably cause this interfacial tension to decrease. Therefore, the angles shown in Fig. 15 are maximum angles, which are true contact angles only if the methane would be completely expelled from the real n -nonane/water interface, in the methane/ n -nonane/water system. We will therefore assume that n -nonane is completely wetting on water. If this is not the case, contact angles will be small for the cases of interest to us. On the contrary, water is (almost) completely nonwetting on n -nonane, making it very unlikely that it will heterogeneously nucleate onto n -nonane.

B. Heterogeneous nucleation

We will now proceed to calculate the differences in nucleation rates for heterogeneous versus homogeneous nucleation, assuming that n -nonane is complete wetting on water. The question then is, for what radius r_w of the water droplets does the rate of heterogeneous nucleation of n -nonane onto water droplets prevail over the homogeneous nucleation of n -nonane.

The rate of homogeneous nucleation is given as

$$J_{hom} = K_{hom} \exp\left(-\frac{\Delta\Omega_{hom}^*}{k_B T}\right), \quad (8)$$

where K_{hom} is a rate factor for homogeneous nucleation and $\Delta\Omega_{hom}^*$ is the work of formation of a critical cluster.¹⁸ It is given by

$$\Delta\Omega_{hom} = -(p_n - p^G)V_n + \sigma_{nm}A_n, \quad (9)$$

in which p_n is the pressure in the n -nonane droplet (or cluster), V_n is its volume, and A_n is its surface area. The pressure difference between the gas phase and the cluster is given by

$$p_n - p^G = \frac{2\sigma_{nm}}{r_n}. \quad (10)$$

Likewise, the rate of heterogeneous nucleation is given as

$$J_{het} = n_w K_{het} \exp\left(-\frac{\Delta\Omega_{het}^*}{k_B T}\right), \quad (11)$$

where n_w is number density of water droplets. This equation only applies when $r_w \leq r_n$. If $r_w > r_n$, we have a process of activation, not heterogeneous nucleation. Each water droplet of $r_w > r_n$ immediately grows by condensation of n -nonane. The grand potential of the system with a water droplet is given by

$$\Omega_w = -p^G(V - V_w) - p_w V_w + \sigma_{wm}A_w. \quad (12)$$

The grand potential of a system with a water droplet coated with n -nonane is

$$\Omega_{wn} = -p^G(V - V_{nw} - V_w) - p_{wn}V_w - p_n V_n + \sigma_{nm}A_n + \sigma_{wn}A_w, \quad (13)$$

where $V_{nw} = V_n - V_w$. The work of formation of the n -nonane coating $\Delta\Omega_{het}$ then results as

$$\Delta\Omega_{het} = \Omega_{wn} - \Omega_w = -(p_n - p^G)V_{nw} - (p_{wn} - p_w)V_w + \sigma_{nm}A_n + (\sigma_{wn} - \sigma_{wm})A_w. \quad (14)$$

The pressure differences are in this case given by the following Laplace equations:

$$p_n - p^G = \frac{2\sigma_{nm}}{r_n}, \quad (15)$$

$$p_{wn} - p_n = \frac{2\sigma_{wn}}{r_w}, \quad (16)$$

and

$$\begin{aligned} p_{wn} - p_w &= (p_{wn} - p_n) + (p_n - p^G) - (p_w - p^G) \\ &= \frac{2(\sigma_{wn} - \sigma_{wm})}{r_w} + \frac{2\sigma_{nm}}{r_n}. \end{aligned} \quad (17)$$

Comparison of the work of formation for heterogeneous nucleation and for homogeneous nucleation then results in

$$\Delta\Omega_{het} - \Delta\Omega_{hom} = \frac{1}{3}A_w(\sigma_{wn} - \sigma_{wm}). \quad (18)$$

Because $\sigma_{wn} < \sigma_{wm}$ (resulting from the condition of complete wetting), the work of formation of the n -nonane coating on a water droplet $\Delta\Omega_{het}$ is always (for any r_n) smaller than the work of formation of a homogeneous nucleated critical n -nonane cluster. However, the direct comparison of nucleation rates is not possible, because the heterogeneous

nucleation rate depends on the number density of heterogeneous nuclei. When the water droplet radius r_w is larger than the critical radius r_n^* , the n -nonane vapor will immediately start to grow onto the water droplet. The process is called activation rather than heterogeneous nucleation. For water droplets that are only a little larger than r_n^* , there will be a transition between the stochastic heterogeneous nucleation and the deterministic onset of droplet growth by activation. For the conditions studied here, the supersaturation of n -nonane is always relatively large ($S_n > 1.5$), resulting in a critical n -nonane cluster which is always relatively small ($r_n^* < 10$ nm).

The process of heterogeneous nucleation of water onto n -nonane is much less probable. In Fig. 15 the contact angle of water on n -nonane is shown for the two pressures. In all cases the contact angle is close to 180° , indicating that water is almost completely nonwetting on n -nonane. When the interfacial tension between n -nonane and water is decreased, the angle becomes even closer to 180° , up to the point where water becomes completely nonwetting on n -nonane.

V. CONCLUSION

From the analysis of the droplet growth results in the binary systems the diffusion coefficients of water in methane and n -nonane in methane have been determined experimentally. The values of these diffusion coefficients compare well to the predictions of the semi-empirical Fuller correlation.

The droplet growth rates are given as a function of the (supersaturated) molar vapor fraction at constant (averaged) pressure and temperature. By extrapolating the experimentally found droplet growth rates to zero, the equilibrium molar vapor fraction at that pressure and temperature can be obtained. It is observed that equilibrium molar vapor fractions, calculated with the CPA equation of state, agree very well with these extrapolations.

When looking at the droplet growth rates in the ternary system, one can distinguish between two situations. In the first situation one starts with pure n -nonane droplets. The additional presence of supersaturated water vapor will then not increase the growth rate of these droplets. Apparently, the energy barrier for heterogeneous nucleation of water vapor onto n -nonane droplets, created by the newly formed interfacial energies, is larger than the energy barrier for homogeneous nucleation of water. In the second situation one starts with pure water droplets. In this case the presence of supersaturated n -nonane does increase the growth rate of the droplets. Now, the heterogeneous nucleation of n -nonane vapor onto water droplets has a smaller energy barrier than the homogeneous nucleation of n -nonane. The growth rate of these multi-component droplets approaches the growth rate of n -nonane droplets as the molar vapor fraction of n -nonane is increased. Since liquid water and n -nonane are nearly immiscible, and the surface energy of liquid n -nonane is smaller than the surface energy of liquid water, we assume that the n -nonane forms a shell around the water core and eventually completely blocks the water growth. The modeling of this layered growth will be the subject of the companion paper.⁴

FIG. 16. The 90° scattering intensity of vertical polarized light for layered droplets with a water core and a n -nonane layer, as a function of the droplet radius.

APPENDIX: LIGHT SCATTERING BY LAYERED DROPLETS

For the binary experiments of n -nonane in methane or water in methane, the formed droplets will be homogeneous and consist (mainly) out of n -nonane or water, respectively. However, for experiments with ternary mixtures of n -nonane and water in methane, we also expect the formation of layered droplets. These droplets will have a core that is rich in water and an outer layer that is rich in n -nonane. In order to approximate the scattering properties of these droplets we will assume them to be spherically symmetric. The scattering properties of these (spherical symmetric) particles will be different for different ratios r_n/r_w , with r_w the radius of the inner water core and r_n the radius of the outer n -nonane layer. Therefore, the scattering and extinction efficiencies were calculated for different ratios of the inner and outer radius. The results are shown in Figs. 16 and 17. For the calculations use was made of the program DMiLay by Wiscombe.²¹ From Figs. 16 and 17 it can be seen that the scattering and the extinction efficiencies of the layered droplets are almost indistinguishable of those of pure n -nonane,

FIG. 17. The extinction efficiency of vertical polarized light for layered droplets with a water core and a n -nonane layer, as a function of the droplet radius.

when the outer radius of the n -nonane layer is twice as large as the radius of the water core. We expect that in most cases the n -nonane layer will largely block the growth of the water core, causing the n -nonane layer to grow much thicker than the radius of the water core. Therefore, we used the scattering efficiencies of pure n -nonane in all experiments where we expected to observe layered droplets.

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