Supporting information for “Near unity mass accommodation coefficient of organic molecules of varying structure”

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Nonane condensational growth model calculations

The condensational growth of nonane was calculated by numerically solving the coupled differential equations for particle mass, temperature at droplet surface and far away from the droplet, the particle concentration and the partial pressure of the nonane vapor. The differential equations were solved using the freely available solver DVODE. The droplet population is assumed monodisperse and no nucleation or chemical reactions are taking place, and the initial droplet radius was set to the value of the first experimentally measured radius. The difference between nonane partial pressure far from the droplet and at droplet surface is the driving force for the mass transport, and the temperature difference between the temperature far from the droplet and at droplet surface is the driving force for heat transport.

The mass flux $I$ from a droplet with radius $a$ is given by

$$I = \frac{4\pi a p M \beta_m D}{RT_\infty} C \ln \left( \frac{1 - \frac{p_{v\infty}}{p}}{1 - \frac{p_{va}}{p}} \right),$$  

where $p$ is the total pressure, $M$ is the molar mass, $\beta_m$ is the transition regime correction factor, $D$ is the diffusion constant, $R$ is the gas constant, $T_\infty$ is the temperature far from the droplet, $p_{v\infty}$ and $p_{va}$ are the partial vapor pressures far from the droplet and at the droplet surface, respectively, and $C$ accounts for the temperature dependence of the diffusion coefficient. $C$ has the form

$$C = \frac{T_\infty - T_a}{T_\infty} - \frac{2 - \mu}{T_{\infty}^{2-\mu} - T_a^{2-\mu}},$$

where $T_a$ is the temperature at the droplet surface and $\mu$ is the parameter that appears in the temperature dependent diffusion coefficient, $D(T) = D_N(p_N/p)(T/T_N)^\mu$. The mass accommodation coefficient enters the equations through the correction factor $\beta_m$, for which we used the Fuchs-Sutugin expression.
The Knudsen number $Kn_m$ is defined as the ratio of the mean free path of the nonane vapor and the droplet radius.

The heat flux from the droplet is given by

$$ Q = 2\pi a(k_a + k_\infty)(T_a - T_\infty)\beta_T + HI, $$  \hspace{1cm} (S4)

where $k_a$ and $k_\infty$ are the thermal conductivities of the gas mixture at the droplet surface and far from the droplet, respectively, $H$ is the specific enthalpy of the nonane vapor and $\beta_T$ is a transition regime correction factor, given by

$$ \beta_T = \frac{1 + Kn_T}{1 + \left(\frac{4}{3} \alpha_T + 0.337\right) Kn_T + \frac{4}{3} \alpha_T Kn_T^2}, $$  \hspace{1cm} (S5)

where $\alpha_T$ is the thermal accommodation coefficient which in the present work is assumed to be 1, and $Kn_T$ is the ratio of the mean free path of the inert gas (air) molecules and the droplet radius.

The droplet is assumed to be in (quasi) equilibrium with the vapor at the surface, and so the vapor partial pressure at the surface is given by

$$ p_{va} = p_{v,sat} \exp\left(\frac{2\sigma M}{RT_a \rho_l a}\right), $$  \hspace{1cm} (S6)

where $\sigma$ is the surface tension, $\rho_l$ is the liquid density and $p_{v,sat}$ is the saturation vapor pressure, for which we used three different values based on parameterizations of the form\(^4\text{--}^6\)

$$ \ln p_{v,sat} = A_1 + \frac{A_2}{T_a - A_3} + A_4 \ln T_a + A_5 T_a. $$  \hspace{1cm} (S7)

The saturation vapor pressure parameter values are collected in Table S1. Corresponding expressions for the latent heat of vaporization were taken from the same sources. Temperature-dependent expressions for various other nonane properties were used as well.
Winkler\textsuperscript{6} were used, while expressions according to Yaws\textsuperscript{7} were used for the specific heat capacity of the liquid and liquid density. The specific heat capacities of the nonane vapor and air were assumed to be temperature-independent.

References

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Table S1: Saturation vapor pressure parameter values, parameterizations according to eq. S7.

|                     | $A_1$     | $A_2$     | $A_3$ | $A_4$     | $A_5$     |
|---------------------|-----------|-----------|-------|-----------|-----------|
| King & Al-Najjar$^a$| 133.672   | -9467.4   | 0.0   | -17.5683  | 0.0152556 |
| Kulmala et al.$^b$  | 48.3313   | -6363.03  | 0.0   | -3.60267  | 0.0       |
| Winkler$^b$         | 98.8613   | -8201.13  | 0.0   | -11.8717  | 0.00903284|
Figure S1: Schematic illustration of the initial setup for the cluster case. 100 incoming molecules with two angles of approach were simulated: 1) molecules sent directly towards the cluster centre of mass and 2) molecules aimed at the circle depicted in the figure. See also the Methods section in the main text.
Figure S2: Naphthalene density profile and the surface/bulk division. Vertical lines denote the locations of the surface/bulk boundary when the location is defined as a fraction of the bulk condensed phase density. Of the studied systems naphthalene has the widest surface.
Figure S3: Nonane density profile (in arbitrary units). The surface peak is due to the surface monolayer of molecules that are oriented along the surface normal. The tightly-packed peak has higher density than the bulk liquid phase.
Figure S4: Density profiles of the dicarboxylic acids.