Re-evaluation of the Pressure Effect for Nucleation in Laminar Flow Diffusion Chamber Experiments with Fluent and the Fine Particle Model

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This study is an investigation of the effect of total pressure on homogeneous nucleation rates of *n*-butanol in helium and *n*-pentanol in helium and argon in a laminar flow diffusion chamber (LFDC). To verify earlier findings, experimental data was re-evaluated using the computational fluid dynamics (CFD) software FLUENT in combination with the fine particle model (FPM) for aerosol dynamics calculations. This approach has been introduced in an earlier paper [Herrmann, E.; Lihavainen, H.; Hyvärinen, A.-P.; Riipinen, I.; Wilck, M.; Stratmann, F.; Kulmala, M. *J. Phys. Chem. A* 2006, *110*, 12448]. As a result of our evaluation, a flaw in the femtube2 code was found which had been used in the original data analysis [Hyvärinen, A.-P.; Brus, D.; Ždímal, V.; Smolík, J.; Kulmala, M.; Viisanen, Y.; Lihavainen, H. *J. Chem. Phys.* 2006, *124*, 224304]. The FLUENT analysis yielded a weak positive pressure effect for the nucleation of *n*-butanol in helium at low nucleation temperatures (265−270 K). *n*-Pentanol in helium showed a positive pressure effect at all temperatures (265−290 K), while the effect for the nucleation of *n*-pentanol in argon was negative at high temperatures (280 and 285 K) and positive at lower nucleation temperatures (265 K). These findings support results gained with the corrected femtube2 model. In this study, we also carried out a detailed comparison of FLUENT and femtube2 modeling results, especially focusing on the calculation of temperature and saturation ratio at nucleation rate maximum (T_{nuc} and S_{nuc}, respectively) in both models.

1. Introduction

In nucleation studies, the term "pressure effect" usually denotes the influence of carrier gas total pressure on the observed nucleation rate. Despite theoretical and experimental investigation, this issue remains still unclear and open.[3] While basic theories such as the classical nucleation theory do not consider the role of a noncondensible gas in nucleation, several experiments and model studies have shown that total pressure can influence the nucleation process.[4] Experimentally, the pressure effect has been repeatedly observed in diffusion-based devices such as the thermal diffusion cloud chamber (TDCC) and the laminar flow diffusion chamber (LFDC), which yield a comparable pressure effect for the *n*-alcohols.[3] However, pressure-dependent devices do not typically yield any pressure effect. From a theoretical point of view, a recent molecular dynamics simulation suggests that both a negative and positive pressure effect may be observed, depending on the nucleation conditions.[5]

Most of the recent experiments concerning the pressure effect have been conducted with a TDCC or a LFDC.[2−4] These experiments have yielded both positive and negative pressure effects. The results show clear relation to both the nucleation temperature and saturation ratio regardless of substance or device. However, the evidence is still indicative rather than conclusive, and reasons related to artifacts need to be ruled out for a better understanding of the results.

One of these is the accuracy of the mathematical model used for calculating the temperature and partial vapor pressure profiles in the LFDC.[3] As these parameters cannot be measured directly, theoretical fluid dynamics models are needed to describe the flow profiles in the chamber. The model used in the original data analysis, femtube2, is an in-house built, simple model[6] that does not take into account any aerosol effects such as condensation of vapor on particles. To judge the accuracy of this model, a more rigorous approach is needed.

FLUENT is a flow simulation software whose complex possibilities are available to aerosol modeling through the recently developed fine particle model (FPM).[7−9] FLUENT-FPM provides full fluid and aerosol dynamics. Because only geometry, materials, and boundary conditions need to be defined, the program is a natural choice to set up an alternative model for error analysis in nucleation measurements. FLUENT has been used previously to analyze results from LFDC measurements conducted at atmospheric pressure, and this work is the continuation of the previous paper.[1]

Here, we study the pressure-dependent nucleation measurements conducted in a LFDC by using FLUENT-FPM as the model to calculate the flow and particle profiles in the LFDC. The intention of the study is to assess the accuracy of the simpler, conventionally used femtube2 model at different pressures. For this, the homogeneous nucleation rate results of *n*-butanol in helium[2,10] and *n*-pentanol in helium and argon[3] at pressures from 50 to 400 kPa are reanalyzed.

2. Materials and Methods Section

2.1. FLUENT and the Fine Particle Model. FLUENT (version 6.3.26, Fluent Inc.) is a commercially available CFD (computational fluid dynamics) software which models flow based on the Euler equations for mass and momentum conser-

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vation. These equations are solved using a control-volume-based technique that divides the simulation domain into discrete control volumes on which the governing equations are integrated to build equations for the discrete dependent variables (velocity, pressure, etc.). These discretized equations are then linearized and solved to obtain updated values of the dependent variables. Because of the nonlinearity of the governing equations, several iterations (depending on the convergence criteria up to several thousand) of the solution loop are necessary to reach convergence. A typical FLUENT simulation calculates the following quantities for each cell of the simulation domain: pressure, density, velocity, temperature, and, in case of multiple species, the mass fraction of each species. Additional quantities can be studied by adding user-defined functions (UDF).[11]

The fine particle model (FPM, version 1.4.2, Particle Dynamics GmbH & Chimera Technologies) is a complex UDF that adds a particle dynamics model to FLUENT. The FPM simulates formation, transformation, transport, and deposition of multicomponent particles in gases and liquids. The applicable size range stretches from molecule size up to micrometer particles. The FPM simulates the dynamics of a particle population, which means that a statistically significant number of particles must exist in the simulation domain, such that their size distribution can be represented by a continuous function. The FPM solves for the spatial and temporal evolution of a multimodal, multiphase, multispecies particle size distribution. In FPM 1.4.2, the particle size distribution is represented by a superposition of log-normal size distribution functions (modes). The particle dynamic equations are solved using the moment method; integral moments of the modes (for example, total number) become additional scalars in FLUENT.

Properties and capabilities of FLUENT and the FPM that are relevant in our simulations as well as some central equations are described in more detail in a previous paper on the subject.[1]

2.2. Thermodynamics of *n*-Butanol and *n*-Pentanol. Generally speaking, the software considers two different species; FLUENT deals with a mixture of nucleating vapor (*n*-butanol or *n*-pentanol in our case) and carrier gas (helium or argon), and the FPM deals with liquid droplets of a nucleated substance. These two are connected by nucleation, condensation, and evaporation in FPM. However, both species have to be defined separately in FLUENT and the FPM. The definitions for all substances and their properties in this work are taken from refs 2 and 3. Depending on their availability, properties can be defined in different ways for different materials. Mostly, those definitions are polynomials or other explicit expressions. In some cases, however, built-in FLUENT functions were used. As the source code of FLUENT is not publicly available, this causes certain “black box” effects, that is, we are not always able to fully determine FLUENT operation.

3. Simulations

The laminar flow diffusion chamber (LFDC) used in the experimental foundation of this work is described in detail in ref 2. Figure 1 shows the basic setup. In the experiment, carrier gas becomes saturated with nucleating vapor in the saturator. The mixture then enters the preheater, whose function it is to ensure well-defined conditions. Upon entering the condenser (at lower temperature than the preheater and saturator), the mixture gets cooled down. Since vapor diffusion is slower than heat diffusion for the materials used in our experiments, the mixture becomes supersaturated, and at sufficiently high values of S, we observe nucleation, that is, the formation of new particles, at the center of the tube. Nucleated particles are counted downstream from the condenser.

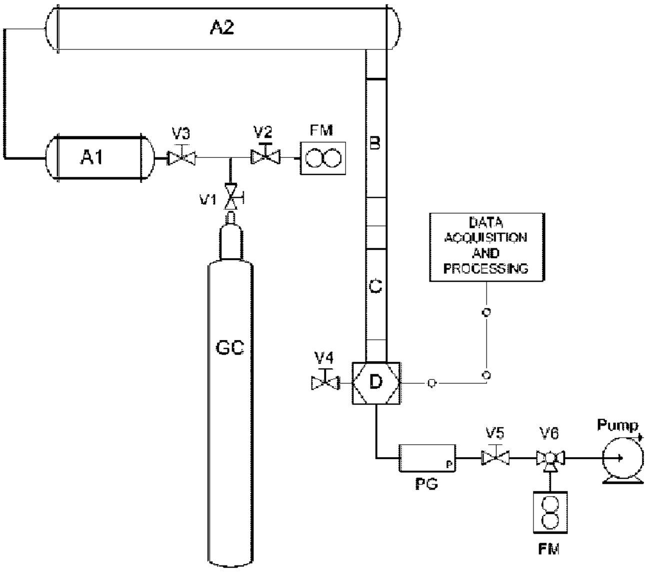

Figure 1. Schematic figure of the experimental setup. A1: presaturator; A2: saturator; B: preheater; C: condenser; D: optical cell; FM: flow meter; GC: gas container; PG: pressure gauge; V1−V6: valves.

In the simulation, this setup needs to be reduced as much as possible to save computing time. Thus, the saturator is included as a boundary condition at the inlet, and the simulated preheater is much shorter than the actual one. This latter change is possible since the flow reaches a steady state already a few centimeters inside of the preheater. The condenser, on the other hand, is a few centimeters longer than its real-life counterpart to prevent simulation boundary effects at the flow exit from affecting our volume of interest.

As in our previous work, the nucleation rate is calculated using the kinetically corrected version of the classical nucleation theory formulation by Becker-Döring.[12] In comparison with our earlier work, we are now using a 2D grid with higher resolution (250000 grid cells). To account for different pressures, the outlet boundary type had to be changed from simple “outflow” to “pressure outlet”. In comparison with the conventionally used femtube2 model, there are two main differences. First, the radial resolution in femtube2 is 14, while the resolution of the grid used in the FLUENT simulations is 200. Furthermore, femtube2 calculates only temperature and vapor profiles, which are then used to determine a nucleation rate. Femtube2 does not take into account the effect of particles on these profiles. The FPM, on the other hand, calculates nucleation rates within FLUENT, and thus, the condensation of vapor on particles is included in the simulation (vapor depletion[1]).

4. Results and Discussion

4.1. Simulation Data. FLUENT-FPM delivers (among others) data sets of flow velocity, saturation ratio, nucleation rate, and particle concentration for the whole simulation domain. However, the maximum experimental nucleation rate $J_{\mathrm{exp}}^{\mathrm{max}}$ is normally used as the resulting nucleation rate from flow chamber experiments and, thus, is the logical choice to compare our simulations and earlier theoretical results. Equation 1 to obtain $J_{\mathrm{exp}}^{\mathrm{max}}$ from a LFDC has been proposed in ref 13

$$\frac{J_{\mathrm{exp}}^{\mathrm{max}}}{\int J_{\mathrm{exp}}\mathrm{d}V} = \frac{J_{\mathrm{theo}}^{\mathrm{max}}}{\int J_{\mathrm{theo}}\mathrm{d}V} \qquad (1)$$

For our purposes, this becomes

$$J_{\mathrm{exp}}^{\mathrm{max}} = J_{\mathrm{theo}}^{\mathrm{max}} \frac{N_{\mathrm{exp}}}{N_{\mathrm{theo}}} \qquad (2)$$

where $J_{\mathrm{theo}}^{\mathrm{max}}$ is the maximum theoretical nucleation rate on the central axis of the simulation tube. N_{exp} is the number concentration measured in the actual experiment, whereas N_{theo} is the theoretical concentration as predicted by a model, in our case, FLUENT-FPM or femtube2. It is the average of particle concentrations (weighed with the flow velocity in each respective cross-section cell) over the condenser outlet.

4.2. Simulation Results. ***4.2.1. Nucleation of n-Butanol in Helium.*** On the basis of experimental data published in ref 2, nucleation isotherms of *n*-butanol in helium were simulated at 50, 100, and 200 kPa for nucleation temperatures (i.e., temperature at the nucleation rate maximum) of 265, 270, and 280 K. The highest, middle, and lowest point of each isotherm were chosen. This way, we could cover the range of experimental data at reasonable computing costs.

Nucleation measurements of *n*-butanol in helium published in ref 2 exhibited a negative pressure effect. Our FLUENT-FPM simulations did not confirm the pressure effect that was reported in 2006. Figure 2 illustrates that while nucleation rates at 50 kPa and 280 K appear left of the 100 kPa ones with femtube2, and FLUENT-FPM simulations show results from both pressures landing on top of each other. A clue for this was found when comparing the results of nucleation rates versus total pressure. For these experiments, the saturation ratio was kept constant according to the femtube2 model (Figure 3). However, with the same boundary conditions, FLUENT-FPM shows noticeably higher saturation ratios at lower pressures (<150 kPa). At high pressures, the difference practically disappears. The nucleation rates showed a similar, decreasing trend with pressure with both models.

These findings prompted a thorough comparison of both models. It showed that femtube2 calculates the heat capacity of the gas−vapor mixture based on mole fractions, while the heat capacities of the single components were defined inconsistently in terms of mass. This was corrected in the femtube2 code, and the corrected results have later been published.[10] The results calculated with the corrected femtube2 and FLUENT-FPM show very closely the same results, as Figure 2 shows.

Figure 4 shows an overview of the results, including corrected femtube2 results for comparison. We see a weak positive effect at nucleation temperatures of 265 and 270 K and no clear effect at 280 K. The positive effect lies within the limits of experimental uncertainty, which was determined in ref 2. The figure also shows different slopes of the isotherms at different pressures. With growing pressure, the slopes decrease. In the figure, this effect is most clearly visible for the 270 K data.

To get a better picture of how femtube2 and FLUENT-FPM work in comparison, we analyzed in detail the predicted nucleation temperatures and saturation ratios. The FPM predicts systematically somewhat lower nucleation temperatures. Depending on the setup, these differences range from 0 to 0.06 K. If we, however, estimate an error value for the FPM's T_{nuc} based on grid resolution and thus exact location of the nucleation maximum, we find that femtube2 T_{nuc} values generally fit well within this uncertainty range.

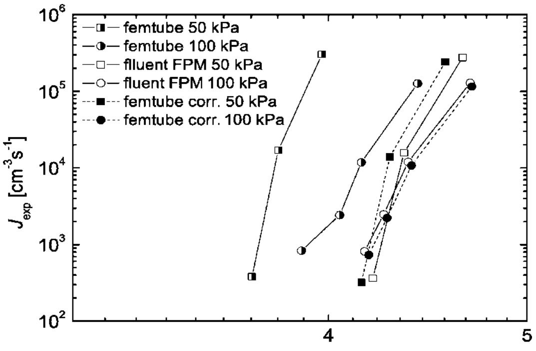

Figure 2. Nucleation rates of *n*-butanol in helium at 280 K at 50 and 100 kPa. Comparison of the FLUENT-FPM analysis with results from the original and the corrected femtube2 model. For all model calculations, the same boundary conditions were used.

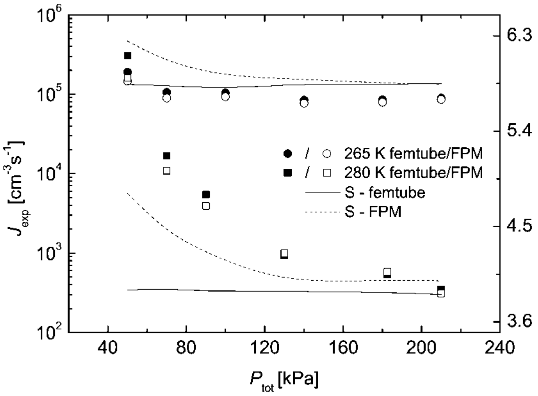

Figure 3. Nucleation of *n*-butanol in helium at 265 and 280 K from 50 to 200 kPa. Comparison of the pressure dependence of the maximum experimental nucleation rate $J_{\mathrm{exp}}^{\mathrm{max}}$ and the saturation ratio *S* given by FLUENT-FPM and by the original, uncorrected femtube2 model using the same boundary conditions.

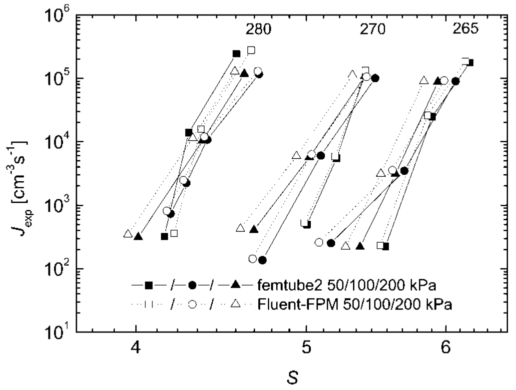

Figure 4. Nucleation rates of *n*-butanol in helium as a function of saturation ratio. Results calculated with FLUENT-FPM and the corrected femtube2 model using the same boundary conditions.

Furthermore, we analyzed the difference in predicted saturation ratios S_{nuc} between both models. The dependence of this ΔS on pressure and nucleation temperature is shown in Figure 5. In the figure, ΔS ranges from −0.1 to +0.1, systematically growing with rising pressure and with decreasing nucleation temperature. These two dependencies can be combined into one; ΔS depends on the amount of vapor in the vapor−gas mixture. With decreasing vapor load (higher pressures, lower temperatures), ΔS approaches a "final" value of about +0.1, which is not anymore significantly affected by the vapor load. This value is probably caused by the mentioned differences between both models, grid resolution and particle feedback on the vapor profile. With *n*-pentanol, we see supporting evidence for this explanation. In the *n*-pentanol case, the vapor mass fraction is generally well below the levels for *n*-butanol, and indeed, we see a practically constant ΔS in the *n*-pentanol case, indicating that the vapor load does not play a significant role. At high

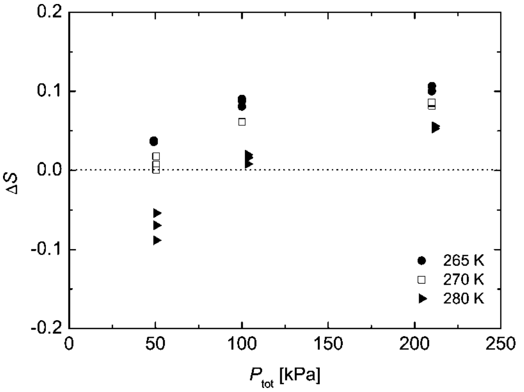

Figure 5. The difference in saturation ratio ΔS (= S(femtube2) − S(FLUENT-FPM)) between the corrected femtube2 and FLUENT-FPM as a function of pressure and nucleation temperature for the nucleation of n-butanol in helium.

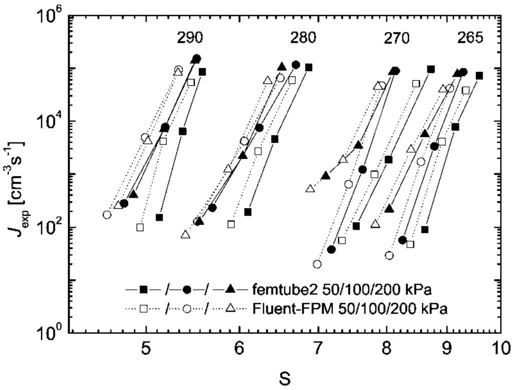

Figure 6. Nucleation rates of n-pentanol in helium as a function of saturation ratio. Results calculated with FLUENT-FPM and the corrected femtube2 model using the same boundary conditions.

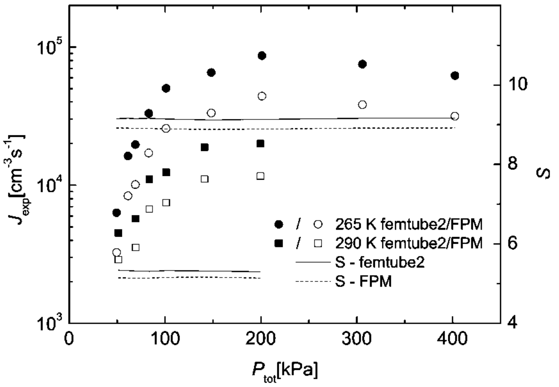

Figure 7. Nucleation rate and saturation ratio as a function of total pressure for the nucleation of n-pentanol in helium. Results are calculated with FLUENT-FPM and the corrected femtube2 model using the same boundary conditions.

vapor load, the condensation of vapor affects the flow in the tube, and the behavior of ΔS in the n-butanol case is a result of how femtube2 and FLUENT take this into account. Femtube2 simply assumes a constant laminar flow field throughout the tube, ignoring the influence of temperature changes and vapor losses on the flow. FLUENT takes these into account. With little or no vapor in the gas, we observe a drop in flow velocity when the temperature drops in the condenser. The ideal gas law and mass conservation demand this. With a high vapor load, the situation becomes more complicated. With the models used in our simulations, FLUENT generally enforces mass conservation. This means that the vapor that condenses to the condenser walls is replaced by carrier gas to keep the total mass constant. At high vapor mass fractions, this leads to a large excess volume of carrier gas in the tube that causes higher flow velocities than a physically correct treatment would yield. This can be corrected in FLUENT, but the methods are costly in terms of computing time. Also, the effect is significant only for a few extreme setups, and even then, the nucleation maximum is reached before the simulated flow velocity reaches even preheater values. As it is, this effect only reduces the "flow velocity step" at the condenser entrance in those cases where the vapor mass fraction is very high. At moderate vapor mass fractions, and in the majority of our simulations, this effect does not play a significant role. Finally, even in the n-butanol case, this effect ($\Delta S = 0.2$ in the most extreme case, below 0.1 for most cases) is smaller than the estimated experimental uncertainty ($\Delta S = 0.35^2$), and thus, costly correction methods were not implemented.

In general, FLUENT-FPM simulations confirm the results of the corrected femtube2 model, as Figure 4 shows. Despite slight systematic differences between both models, the overall findings and tendencies of the corrected femtube2 analysis[10] are supported by FLUENT.

4.2.2. Nucleation of n-Pentanol in Helium. On the basis of experimental data published in ref 3, nucleation isotherms of n-pentanol in helium were simulated at 50, 100, and 200 kPa for nucleation temperatures of 265, 270, 280, and 290 K. Also for n-pentanol and helium, the agreement between FLUENT-FPM and femtube2 is very good. While there are small systematical differences between single data points, FLUENT-FPM supports the general findings of the femtube2 analysis. Figure 6 shows an overview of the results, including femtube2 results[3] for comparison. For this work, the femtube2 code was already corrected. At all nucleation temperatures, a positive pressure effect can be seen; the isotherms at lower pressure are at higher values of S than the isotherms at higher pressure. As in the n-butanol case, isotherm slopes depend on pressure; with increasing pressure, isotherm slopes decrease.

Additionally, some series of measurements were made to study the dependence of the nucleation rate on pressure, while the saturation ratio was kept constant. Figure 7 shows the analysis of these data sets at the minimum and maximum nucleation temperatures of 265 and 290 K. In this figure, we clearly see that nucleation rate increases with growing pressure. This time, the saturation ratio with both models has the same value. The effect is most prominent below pressures of 100 kPa and levels out beyond 200 kPa.

A detailed comparison of FLUENT and femtube2 results shows the saturation ratios in FLUENT to be 0.15−0.20 below the respective femtube2 values. A systematic dependency of this ΔS on nucleation temperature or pressure cannot be seen. As previously stated,[1] this difference originates in the definition of the diffusion coefficient for n-pentanol in helium through the built-in "kinetic theory" model in FLUENT, whose operation cannot be completely determined by the user. Comparing T_{nuc} values in FLUENT and femtube2 did not yield any significant difference between the models. With similar uncertainty limits as thosedefined above for the n-butanol case, the difference between FLUENT and femtube2 (from −0.04 to 0.04 K) is generally smaller than the respective error margin (up to 0.1 K).

4.2.3. Nucleation of n-Pentanol in Argon. Finally, the nucleation of n-pentanol in argon was simulated, also based on experimental data published in ref 3. Since all simulations so far have shown good agreement between femtube2 and

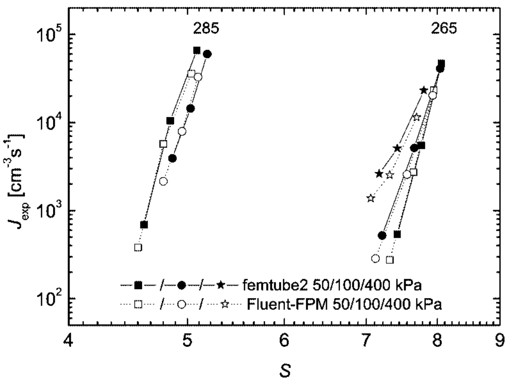

Figure 8. Nucleation rates of *n*-pentanol in argon as a function of saturation ratio. Results calculated with FLUENT-FPM and the corrected femtube2 model using the same boundary conditions.

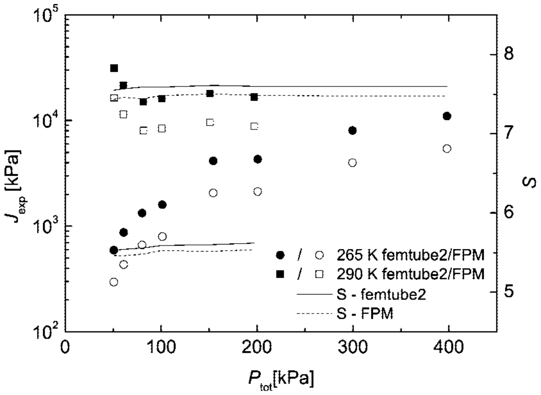

Figure 9. Nucleation rate and saturation ratio as a function of total pressure for the nucleation of *n*-pentanol in argon. Results are calculated with FLUENT-FPM and the corrected femtube2 model using the same boundary conditions.

FLUENT-FPM, we simulated fewer data points. At 265 and 285 K nucleation temperatures, isotherms were simulated for 50 and 100 kPa pressures. Additionally, at 265 K, one isotherm was calculated for 400 kPa. Figure 8 shows the results. Results derived with both models agree exceptionally well. At 265 K, we see a positive pressure effect, while at 285 K, there appears to be a negative effect. As in all other studied cases, isotherm slopes decrease with increasing pressure.

Similar to the *n*-pentanol−helium case, we also studied the dependence of the nucleation rate on pressure at a constant saturation ratio for two different nucleation temperatures, 265 and 280 K. Figure 9 shows the results. In agreement with our interpretation of Figure 8, this figure shows a small negative pressure effect at 280 K. At lower temperatures, we observe a clear positive effect. Other than with helium as the carrier gas, this positive effect continues well over the 200 kPa mark up to 400 kPa (and probably beyond). Reference 3 explained this with the adsorption of argon on the droplet surface.

Comparing FLUENT and femtube2 results in the argon case, we found FLUENT's saturation ratios to be less than 0.1 smaller than the femtube2 values. We did not observe a systematic dependency on pressure or nucleation temperature. Compared to the helium simulations, this is the "cleanest" difference between the models, that is, not caused by one major flaw of either model but instead the sum of all of the differences between the models, from grid resolution to solution algorithm. For argon, differences in T_{nuc} are somewhat larger than those in the helium simulations, about as large as the uncertainty limits (up to 0.1 K). However, T_{nuc} is systematically smaller in FLUENT, which indicates an actual, if small, difference between both models. That this difference can be seen only for argon is most likely caused by the fact that the nucleation maximum in argon simulations occurs much later (in time) than that in the helium case. Possibly existing differences have thus more time to grow and become observable.

5. Conclusions

Nucleation in a laminar flow diffusion chamber was simulated with FLUENT-FPM in order to re-evaluate experimental data and verify previous findings. Nucleating species were *n*-butanol and *n*-pentanol, and helium and argon were used as carrier gases. Pressures ranged from 50 to 400 kPa, and nucleation temperatures ranged from 265 to 290 K.

For the nucleation of *n*-butanol in helium, a weak positive effect was found at the lower end of the nucleation temperature range (265−270 K). No clear effect can be seen at higher temperatures. During data analysis, a mistake was found in the earlier femtube2 code, which had lead to an apparent, but false, negative effect. The error was analyzed and quantified.

The nucleation of *n*-pentanol in helium showed a clear positive pressure effect at all nucleation temperatures from 265 to 290 K. The pressure effect can only be seen at lower pressures; over 200 kPa, it flattens out. In the case of *n*-pentanol nucleation in argon, we found a small negative effect at higher temperatures (280 K), while the effect is clearly positive at low nucleation temperatures (265 K). Other than in the helium case, the effect continued at higher pressures beyond 200 kPa.

Generally speaking, our simulations support the results obtained with the corrected femtube2 model, previously reported in refs 3 and 10. This means that also the use of a more accurate mathematical model yielded a pressure effect. Negative and positive pressure effects were found, depending on the nucleation temperature and carrier gas. This suggests two competing mechanisms that affect the nucleation process, with the negative effect being amplified by a lighter vapor, a heavier carrier gas, and higher temperatures. According to a recent publication,[5] the effect (at low pressures) arises from a competitive effect of nonisothermal nucleation and the extra work that a growing cluster has to do against the pressure of the carrier gas.

Results from FLUENT-FPM simulations and femtube2 model calculations were compared with special emphasis on their prediction of the saturation ratio and nucleation temperature. Between both models, saturation ratio values essentially differ only within experimental uncertainty limits, as established in ref 2. On this small scale, however, systematic differences can be found, most prominently for the nucleation of *n*-pentanol and *n*-butanol in helium. In terms of nucleation temperature, FLUENT-FPM and femtube2 predict very similar results. In the most extreme cases, the difference between both models is on the order of 0.1 K, and even in these cases, the difference is not larger than estimated FLUENT-FPM error margins based on grid resolution. The most significant deviation that we found was for the nucleation of *n*-pentanol in argon, where ΔT_{nuc} is still within error margins but suggesting slight systematic differences. Generally, FLUENT-FPM results agree very well with the femtube2 analysis. Under the given experimental conditions, potentially problematic effects such as vapor depletion[1] do not play a significant role.

In this work, we have improved our knowledge of the carrier gas pressure effect by first identifying an error in the previous model and subsequently verifying the results of the corrected model with a more sophisticated approach, as suggested in ref

3. A detailed comparison of the results obtained with both models shows that, despite small systematic differences, deviations between both models do not outgrow experimental uncertainty limits. Beyond this work, more measurements are needed to further investigate the relationship between vapor properties and the pressure effect. Additionally, a theoretical analysis of the pressure effect at conditions similar to the ones in the experiment is needed.

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