

D₂O-H₂O condensation in supersonic nozzles.

II. Modeling

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Abstract. An integral steady state model of nucleation and condensation was used to examine the formation and growth of D₂O droplets in a supersonic nozzle. The classical nucleation rate expression was used together with isothermal and nonisothermal droplet growth laws. For each experiment, the nucleation rate expression was multiplied by a temperature independent parameter in order to match the experimentally observed onset of condensation. In all cases, the predicted pressure traces lie above the measured ones. For one of the condensation experiments, the corresponding neutron scattering spectrum was also available. Modeling showed that once the rate expression was adjusted to match onset, the predicted scattering spectrum was a strong function of the growth law. Furthermore, the match between the measured and predicted scattering spectra was much better for the isothermal growth law than for the nonisothermal growth law.

INTRODUCTION

Modeling of condensation in supersonic nozzles has often proceeded concurrently with experiments¹ as a means of inferring nucleation rates, particle concentrations, and particle size distributions. The main problem with this approach is that there is usually no unique pair of nucleation and growth models that gives a best fit to the measured flow pressure² and density³ profiles. Light scattering experiments⁴ provide only limited help in resolving this lack of uniqueness because in the Rayleigh regime the angle-independent scattering signal gives no information about the differential particle size distribution. In our investigations of condensation in supersonic nozzles, we combine conventional pressure trace measurements⁵ with small angle neutron scattering (SANS) experiments^{6,7}. Since the typical wavelength (0.5-1.5 nm) of cold neutrons is smaller than the mean particle radius, SANS spectra are sensitive to the particle size distribution. Thus, independent estimates of the average particle size, $\langle r \rangle$, the width of the size distribution, σ , and the droplet number concentration, N , are obtained using only a weak assumption about the general shape of the distribution (e.g. Gaussian or log-normal). A brief description of the experimental setup and procedures used for this work is presented in an accompanying paper⁸. Here we describe the model applied to predict condensation in our nozzle, and summarize our first attempts to reconcile the pressure trace and SANS data using this model. In this paper only D₂O results are presented.

MODELING

Our model uses the integral steady state approach first developed by Oswatitsch¹ and applied extensively by others²⁻⁴. The model simulates particle nucleation and growth and includes the effects of heat addition to the flow through the diabatic gasdynamics equations². Cluster sizes are assumed to change only by monomer addition and evaporation. The change in the number density, ΔN , of new particles formed at each step is computed from the steady state nucleation rate, J , and the conservation law, $\Delta N = (J/u)\Delta x$, where u is the local flow velocity. The condensate mass fraction, g , can be calculated as a function of position using an appropriate droplet growth law. From g and the latent heat of condensation, ΔH_{vap} , the change in the flow properties can be obtained by integrating the diabatic flow equations using the measured nozzle profile.

In our calculations we used the classical nucleation rate expression, J_{cl} , for J . Following conventional practice, a temperature independent, multiplicative adjustment factor, Γ , is used to bring the calculated and measured values of the onset temperature into agreement; i.e. $J = \Gamma J_{cl}$. The following expression was used for J_{cl} ,

$$J_{cl} = \left(\frac{2\gamma\mu_c}{\pi N_A} \right)^{1/2} \left(\frac{p_v}{k_B T} \right) \frac{1}{\rho_c} \exp \left[-\frac{16\pi}{3} \left(\frac{\gamma}{k_B T} \right)^3 \left(\frac{v_c}{\ln S} \right)^2 \right].$$

Here γ , μ_c , v_c , and ρ_c are the surface tension, molecular weight, molecular volume, and density of the condensate, respectively; p_v is the partial pressure of the condensable vapor, N_A is Avogadro's number, T is temperature, S is supersaturation, and k_B is the Boltzmann constant. The physical property correlations for D₂O were as follows: vapor pressure comes from Hill *et al.*⁹, ΔH_{vap} is derived from the vapor pressure correlation, surface tension and density are those developed by Wölk and Strey¹⁰. The isothermal droplet growth law is that developed by Wegener *et al.*¹¹ with the droplet temperature equal to the local gas temperature. The nonisothermal droplet growth law is that of Peters and Paikert¹². In both cases the mass accommodation coefficient equals one.

Figure 1 illustrates the conventional modeling results. Once the growth law is selected and Γ is adjusted to match onset, there is only a slight difference between the pressure traces for the two growth rate models. Both models overpredict the final pressure ratio, because each predicts that about 90% of the material condenses by the end of the nozzle. In contrast, by integrating the diabatic flow equations⁵ using the measured pressure traces we find that only ~75% of the material condenses. The values of Γ required to make the growth models match the data are included in the legend. These values are not constant, since the temperature dependence of the nucleation rate is overpredicted by classical theory. The predicted pressure traces do not fit the data very well beyond the onset of condensation, although more of the observed pressure trace could be matched when the isothermal growth law was used.

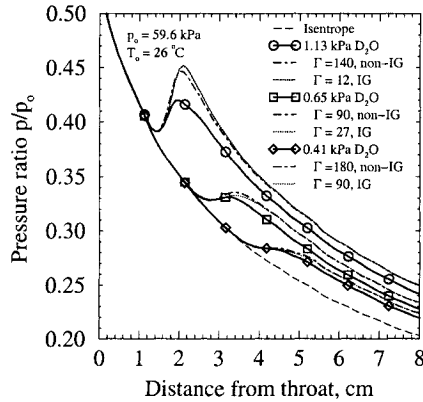


FIGURE 1. Three experimentally measured pressure traces are compared to isothermal (IG) and nonisothermal (non-IG) growth models. Once a reasonable value of Γ is found, there is no significant difference between predictions of the two growth models; the onset criteria of 0.5 K difference between the condensing and isentropic expansion is predicted within 0.7 mm from where it is observed.

The more interesting results are illustrated in Fig. 2 where we compare the predicted and measured (SANS) scattering spectra and their respective underlying size distributions. As illustrated in Fig. 2(a), the scattering spectrum predicted using the isothermal growth law has a shape similar to that of the measured spectrum. In contrast, the scattering spectrum predicted by invoking nonisothermal growth clearly fails to describe the data. This result is surprising because nonisothermal calculations predict that the growing droplets are significantly hotter than the background gas and, thus, nonisothermal effects should be important.

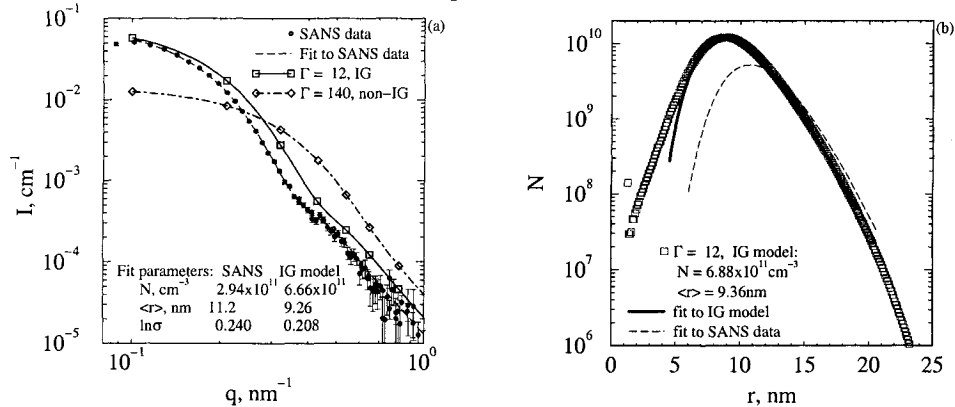


FIGURE 2. 1.13 kPa D_2O results: (a) The measured scattering spectrum, which has been Doppler corrected for a particle velocity of 435 m/s, is compared to the model generated spectra assuming isothermal (IG) or nonisothermal (non-IG) growth. The size distribution parameters for the fit to the SANS and to the IG spectra are given in the legend. (b) The size distribution for the IG model is compared to the log-normal fits to the model generated and measured (SANS) data.

To further quantify the differences between the measured and predicted scattering spectra we first treated the I vs. q data generated by the model using our standard fitting procedures. We found that this synthetic spectrum was far more consistent with

a log-normal particle size distribution than a Gaussian one. As illustrated in Fig. 2(b), the full size distribution predicted by the model is accurately described by a log-normal size distribution for $r > 6$ nm. Both are quite different from the size distribution obtained from the measured SANS spectra. The difference between the SANS and model size distributions indicate that experimentally we observe a smaller number of small particles but roughly the same number of large particles. The larger mean radius of the measured size distribution may indicate that our model growth law underpredicts the actual droplet growth rate. To match the onset of condensation, the modeling appears to make up for the predicted smaller particles by boosting the nucleation rate. As a result, experimentally we observe a narrower size distribution of droplets with larger $\langle r \rangle$ than is modeled. We also compared the experimental and modeled volume fractions of the condensed material. From SANS spectra the deduced condensed volume fraction is $\phi = 2.1 \times 10^{-6}$. The pressure trace data give the same result to two significant figures. The isothermal growth model predicts ϕ of 2.8×10^{-6} , while the fit to the generated $I(q)$ spectra leads to $\phi = 2.9 \times 10^{-6}$. The values of ϕ are consistent with the earlier conclusion that less material actually condenses than is predicted by the model.

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REFERENCES

1. Oswatitsch, K. *Z. Angew. Math. Mech.* **22**, 1 (1942).
2. Hill, P.G., *J. Fluid Mech.* **25**, 593 (1966).
3. Wyslouzil, B.E., Wilemski, G., Beals, M.G., and Frish, M.B., *Phys. Fluids* **6**, 2845 (1994).
4. Moses, C.A. and Stein, G.D., *J. Fluids Eng.* **100**, 311 (1978).
5. Wyslouzil, B. E., Heath, C. H., Cheung, J. L., Wilemski, submitted, (2000).
6. Wyslouzil, B.E., Cheung, J.L., Wilemski, G., and Strey, R., *Phys. Rev. Lett.* **79**, 431 (1997).
7. Wyslouzil, B.E., Wilemski, G., Cheung, J.L., Strey, R., Barker, J., *Phys. Rev. E* **60**, 4330 (1999).
8. Heath, C.H., Streletzky, K.A., Wölk, J., Wyslouzil, B.E., and Strey, R., ICNAA (2000).
9. Hill, P.G. and MacMillan, R.D.C., *Ind. Eng. Chem. Fundam.* **18**, 412 (1979).
10. Wölk, J. and Strey, R., "The Homogeneous Nucleation of Water: A Comparative Study of H₂O and D₂O", presented at the APS meeting in Los Angeles, March 16-20 (1998).
11. Wegener, P.P., Clumpner, J.A., and Wu, B.J.C., *Phys. Fluids* **15**, 1869 (1972).
12. Peters, F. and Paikert, B., *Int. J. Heat Mass Transfer.* **37**, 293 (1994).

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