1-1-2000

Thermal diffusion cloud chamber - new criteria for proper orientation

Richard H. Heist
Fairfield University, rheist@fairfield.edu

Daniel Martinez

Yuk Chan

Anne Bertelsmann

https://doi.org/10.1063/1.1361866

Repository Citation

Heist, Richard H.; Martinez, Daniel; Chan, Yuk; and Bertelsmann, Anne, "Thermal diffusion cloud chamber - new criteria for proper orientation" (2000). *Engineering Faculty Publications*. 148.
https://digitalcommons.fairfield.edu/engineering-facultypubs/148

Published Citation

R.H. Heist, D. Martinez, Y. Chan, and A. Bertelsmann. (2000). Thermal diffusion cloud chamber - new criteria for proper orientation. Proceedings of the 15th International Conference on Nucleation and Atmospheric Aerosols 2000, Vol. 534(1), pp. 280-283. American Institute of Physics, Melville, NY. https://doi.org/10.1063/1.1361866

Thermal Diffusion Cloud Chamber - New Criteria for Proper Operation

Richard H. Heist*, Daniel Martinez, Yuk Chan, and Anne Bertelsmann†

Nucleation Laboratory, Department of Chemical Engineering
University of Rochester, Rochester, NY 14627-0166

Abstract. We report results of new nucleation experiments involving 1-pentanol with hydrogen as the background gas utilizing the high-pressure diffusion cloud chamber (HPCC). We discuss the important issue of buoyancy-driven convective motion and cloud chamber operation, and we focus on the lower total pressure limit required for stable chamber operation. We provide, for the first time, an empirical procedure for determining the lower total pressure limit.

INTRODUCTION

The thermal diffusion cloud chamber (TDCC) has been used for nucleation research for more than four decades. It has been responsible for approximately 50% of all critical supersaturation and nucleation rate data published in the literature.[1] The mass and energy transport that occur within the TDCC during operation have routinely been modeled as 1-D transport through a stagnant gas. However, results of recent investigations clearly indicate the need for a careful re-examination of these assumptions.[2-7] In this paper we address the important issue of the lower limit of total pressure for the TDCC. We employ our HPCC with 1-pentanol as our working fluid and hydrogen as our background gas.

EXPERIMENTAL RESULTS

Our recent experiments on the pentanol/hydrogen system span a total pressure range of 10 to 3000 kPa and a nucleation temperature range from 280 K to 380 K. Here, we present only select results of these studies.[8] In Figure 1 we show S_{cr} (critical supersaturation) data as a function of total pressure obtained at constant nucleation temperatures of 331K, 306K, and 280K. The solid line running though each set of data is a best-fit line to the solid data symbols. All open symbols in Figure 1 refer to operational regions of the HPCC that are expected to give rise to unstable behavior and, hence, unreliable data. These data points are discussed below.

In Figure 2 we plot the upper plate temperature for each experiment versus the total pressure corresponding to that same experiment. The upper solid curve is the upper total pressure stability limit for our experiments.[7] The lower solid curve is an empirically determined locus of points that represents the lower total pressure stability limit and will be discussed in more detail below. The solid lines drawn through the nearly vertical data sets are best-fit lines to indicate trends.

CP534, *Nucleation and Atmospheric Aerosols 2000: 15th Int.'l Conf.*, edited by B. N. Hale and M. Kulmala

REGIONS FOR DIFFUSION CLOUD CHAMBER OPERATION

The data presented in Figure 1 represented as solid symbols are limited to those points that were obtained in what we define to be the "proper" operating range for the TDCC. In Figure 2 we see that there are three regions that can be identified that pertain to the reliability of the nucleation data shown in the figures. These three regions are marked I, II and III.

Region III: This region corresponds to a range of undesirable total pressures. When the total pressure in the diffusion cloud chamber is increased above a certain limit, local inversions of the density gradient occur at the wall of the chamber. These inversions can lead to a buoyancy-driven convective flow that is extremely difficult to detect and can seriously affect the reliability of the nucleation data.[3-5,7]

Region II: This region corresponds to the range of chamber operation that we define to be "proper." It corresponds to the range of data represented by the solid points in Figure 1. It is in this region that mass and energy transport are presumed to occur as diffusion and conduction through a (supposed) stagnant background gas and to be accurately described by the routinely used heat and mass transport equations.[2-7]

Region I: As seen in Figure 1, at lower total pressures the measured supersaturation tends to deviate from that expected based upon extrapolation of the data in region II. As with previous results,[3,7] we are led to the conclusion that this deviation is caused by a change in the transport process within the cloud chamber. If we consider the constant nucleation temperature data plotted in Figure 2 and linked by solid lines, we note a sudden change in slope at the lower total pressure end of each series. Based upon our experience with operational stability of the TDCC, we conclude that this change in slope is caused by the onset of an additional mode of transport not considered in our analysis of cloud chamber conditions and for this reason we consider data obtained over this range of low total pressures to be unreliable. We have indicated these data points with open symbols on the graph. We have connected these data points using a separate line, and the lower solid line in Figure 2 connects the intercepts of these lines and thus represents the empirically determined boundary between the regions of "proper" chamber operation (i.e. region II) and the (to be avoided) low pressure region of operation (i.e. region I).

We examined the lower total pressure limit of TDCC operation in a more quantitative fashion by examining the conditions that could give rise to an inversion in the density gradient at the central portion of the TDCC in the region just above the lower plate. We have determined that all the data discussed in this paper are stable in this regard.[8] We have also considered the possibility that "double-diffusive" phenomena may be responsible for this behavior.[9] Application of the criteria for bounds of double-diffusive phenomena to our experimental data, however, did not support this explanation. It may also be that Bénard or Marangoni instabilities in the liquid pool give rise to turbulent flow or mixing in the region just above the surface of the pool that affects the overall transport through the HPCC.[10]

If we examine the stability diagram shown in Figure 2, we make the important observation that at lower temperatures total pressures, the allowable, stable range of operation (region II) becomes seriously limited, and it is increasingly difficult to use the TDCC and obtain reliable nucleation data under these conditions.

We do not yet have an analytical expression to predict this lower total pressure limit, but we can determine this lower limit empirically.[8] The supersaturation is not the only quantity that shows a sudden change in behavior at the lower total pressure boundary. In fact, the heat flux, the mass flux and the plate temperatures all display a similar behavior. We propose that a combination of all these quantities be used to identify empirically the location of the lower total pressure limit of operation in order to create the stability diagram necessary to determine the limits of "proper" operation for the TDCC.[8]

DETERMINING THE "PROPER" RANGE OF OPERATION

In the past, ratios in excess of two or three of the total pressure to the equilibrium vapor pressure of the working fluid at the lower plate temperature have been used to define stable TDCC operation at lower total pressures.[8] In our study, we found no correlation between the pressure ratio and our empirically determined lower total pressure limit. In fact, we have obtained data points with pressure ratios of approximately 1.5 which lie above the lower pressure limit[11] and data points with pressure ratios of approximately 8 to 10 which fall below the lower limit.[8]

When using the TDCC for nucleation measurements the first step in determining the range of operation is to calculate the upper pressure limit.[7] Next, we recommend a series of S_{cr} measurements at constant nucleation temperature, spanning the largest possible total pressure range consistent with the upper total pressure limit. Extrapolation of the pressure dependent data to lower pressures in conjunction with the heat and mass flux and plate temperature criteria described earlier should be used to empirically determine the lower total pressure limit. The region between the upper and lower total pressure limits gives the range of "proper" TDCC operation.

SUMMARY AND CONCLUSIONS

We report results of new nucleation experiments involving 1-pentanol with hydrogen as the background gas. We discuss the existence of a lower total pressure limit for operating the TDCC, and we describe how violating this limit is manifested by the experimental data. We also provide, for the first time, an empirical procedure for determining the lower total pressure limit. Since the upper and lower total pressure limits together impose substantial limitations on the allowable operational range of the TDCC - especially at lower nucleation temperatures and total pressures - there appears to be a serious question concerning the reliability of a significant amount of the nucleation data in the literature resulting from past TDCC studies. Finally, we suggest that the TDCC is best suited for nucleation measurements at higher temperatures and total pressures where the region of stable operation (e.g., region II) is widest.

* Current address: School of Engineering, Manhattan College, Riverdale, NY 10471
† Current address: Bayer Corporation, Baytown, TX

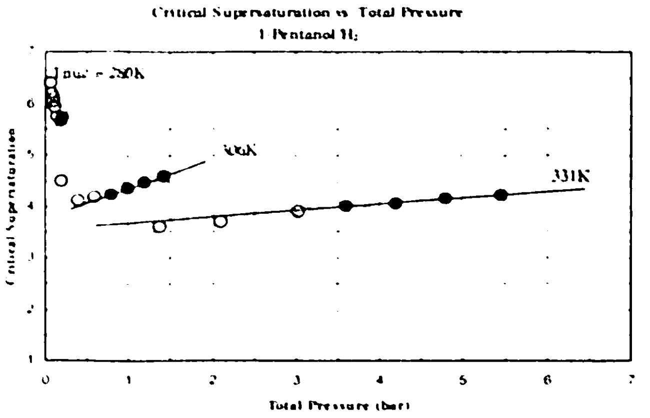

FIGURE 1. Experimentally determined variation of the critical supersaturation of 1-pentanol with total pressure using hydrogen as a background gas. The nucleation temperatures are indicated. The solid lines are fit to the data indicated by solid symbols. See the text for a discussion of the open symbols

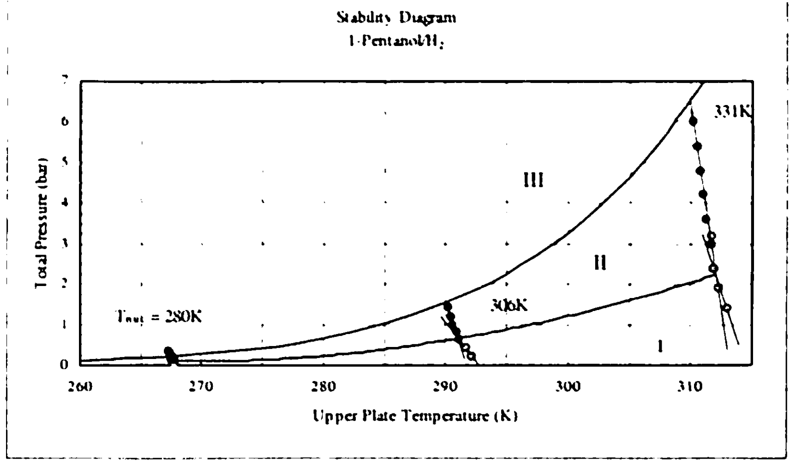

FIGURE 2. Stability Diagram for 1-Pentanol - Hydrogen. The upper and lower total pressure limits of stability are indicated by the solid curves. Data points obtained in the region of "proper" chamber operation (II) are indicated by solid symbols and data points obtained while operating below the lower limit (I) by open symbols. The straight lines are added to illustrate the change in slope of the constant nucleation temperature data.

REFERENCES

1. Heist, R.H. and He, H., *J. Phys. Chem. Ref. Data* **23**, 781 (1994)
2. Bertelsmann, A., Stuczynski, R. and Heist, R.H., *J. Phys. Chem.* **100**, 9762 (1996)
3. Bertelsmann, A. and Heist, R.H., *Atmospheric Research*, **46**, 195 (1998)
4. Ferguson, F.T., Heist, R.H. and Nuth, III, J.A., *J. Chem. Phys.* submitted (2000)
5. Ferguson, F.T. and Nuth, III, J.A., *J. Chem. Phys.* **111**, 8013 (1999)
6. Bertelsmann, A. and Heist, R.H., *J. Chem. Phys.* **106**, 610 (1997)
7. Bertelsmann, A. and Heist, R.H., *J. Chem. Phys.* **106**, 624 (1997)
8. Heist, R.H., Martinez, D., Chan, Y.F and Bertelsmann, A., *J. Phys. Chem.* submitted (2000)
9. Turner, J.S., *Ann. Rev. Fluid Mech.* **17**, 11 (1985); Veronis, G., *J. Mar. Res.* **23**, 1 (1965)
10. Palmer, H.J., private communication.
11. Ye, P., Bertelsmann, A. and Heist, R.H., submitted to *J. Phys. Chem.* (2000)