International Journal of Heat and Mass Transfer 127 (2018) 740–750

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Differential heat and mass transfer rate influences on the activation efficiency of laminar flow condensation particle counters

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ARTICLE INFO

Article history:
Received 7 April 2018
Received in revised form 29 June 2018
Accepted 1 July 2018

Keywords:
Condensation particle counters
Lewis number
Gas mixture transport
Peclet number

ABSTRACT

Laminar flow condensation particle counters (CPCs) are uniquely sensitive detectors for aerosol particles in the nanometer size range (i.e. below 10 nm in size they can have single particle sensitivity). Their operation hinges upon the creation of supersaturation of a working fluid; particles exposed to supersaturated vapor grow by condensation to optically detectable sizes. The degree of supersaturation is fully controlled via differential rates of heat transfer and working fluid vapor mass transfer. Because of the Kelvin relationship governed vapor pressure of small particles, in all CPCs there is a critical size/cut-size (diameter), and particles smaller than this size do not grow and are not detected efficiently. While efforts have been made to control the CPC activation efficiency (i.e. the fraction of particles detected as a function of size), prior studies have not examined how differential heat and mass transfer in CPCs are governed by changes in gas composition. Here, we measure and model CPC activation efficiencies (with 1-butanol as the working fluid) in mixtures of gases of disparate thermophysical properties, namely helium and molecular nitrogen. Our experiments show that the activation efficiency of smaller particles (i.e. below 8 nm in the tested CPC) can be increased by adding a modest amount of helium to the aerosol (mole fractions near 0.20). This is expected based upon the increased Lewis number brought about by Helium addition, and supported by predictions of CPC activation efficiency based upon thermophysical property variable models of coupled heat, mass, and momentum transfer within the CPC condenser region. Interestingly, we find that when operating with a constant precision orifice diameter (choked flow), the activation efficiency for a given sub-10 nm particle diameter first increases with increasing Helium mole fraction and then decreases as the Helium mole fraction increases beyond 0.67. In comparison, experiments with constant mass transfer Peclet number (Pe_m = 77) show an increase in CPC activation efficiency up to a helium mole fraction of 0.67, but then the activation efficiency decreases more modestly beyond this helium mole fraction. We attribute these contrasting results to the increased flowrate through the instrument under constant orifice diameter conditions, which affects the performance of the CPC saturator. Finally, through modeling we show that the ability to enhance the activation efficiency of a CPC via a modest amount of helium addition is general, and can be applied with other heavy working fluids. The results presented in this study elucidate the importance of gas composition and Lewis number controlled differential heat and mass transfer rates on the performance of condensation based nanoparticle detectors.

1. Introduction

The condensation of supersaturated vapor onto nanoparticles and ions in the gas phase has long been utilized as a measurement method to determine nanoparticle number concentration, dating to the work of Aitken [1], Wilson [2–4], and Pollak [5]. In condensation based detection, the supersaturation of a vapor (termed the working fluid) is controlled, such that the vapor will condense onto nanoparticles, while the rate of homogeneous nucleation remains small [6]. Fast condensation growth kinetics result in nanoparticles growing to supermicrometer sizes, which can enable optical detection of individual particles. This process is utilized in a variety of commercially available condensation particle counters (CPCs). There is a wide range of applications of CPCs, including ambient new particle formation monitoring [7,8], vehicle emissions monitoring [9,10], nanoparticle concentration monitoring in clean rooms [11], respirator fit testing [12], and nanoparticle exposure

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https://doi.org/10.1016/j.ijheatmasstransfer.2018.07.002
0017-9310/

Nomenclature

c	concentration of working fluid [mol m^{-3}]
C_p	specific heat at constant pressure [J kg^{-1} K^{-1}]
$\widehat{C}_p$	molar specific heat at constant pressure [J mol^{-1} K^{-1}]
d_c	critical particle diameter [m]
d_p	particle diameter [m]
$D_{\alpha\beta}$	binary diffusivity [m^2 s^{-1}]
k	thermal conductivity [W m^{-1} K^{-1}]
k_B	Boltzmann constant [J K^{-1}]
Le	Lewis number, $k/\rho\ C_p\ D_{AB}$
m	mass of molecule [kg]
M	molecular weight [g/mol]
p	pressure [N m^{-2}]
Pe_m	mass transfer Peclet number, uL/D_{AB}
Pr	Prandtl number, $\mu\ C_p/k$
R	universal gas constant [J mol^{-1} K^{-1}]
Re	Reynolds number, $Lu\rho/\mu$
S	saturation ratio
T	temperature [K]
$\boldsymbol{u}$	gas velocity [m s^{-1}]
v_m	molecular volume [m^3]
x	species mole fraction

Greek letters

α, β	species indices
γ	working fluid surface tension [N m^{-1}]
ε	Lennard-Jones energy parameter [J]
η_{CPC}	CPC activation efficiency
θ	dimensionless temperature
μ	gas viscosity [kg m^{-1} s^{-1}]
ρ	gas density [kg m^{-3}]
σ	Lennard-Jones size parameter [m]
τ_V	DMA transfer function
$\phi_{\alpha\beta}$	dimensionless quantities in viscosity and thermal conductivity mixing rules
χ	helium mole fraction
Ω_D	collision integral for diffusivity
Ω_μ	collision integral for viscosity

Subscripts

b	1-butanol
in	saturator conditions
He	helium
N_2	nitrogen

assessment in industrial settings [13–15]. Given their widespread use, careful characterization of the capabilities and limitations of CPCs is paramount.

Though only investigated in limited circumstances [16–21], CPC performance is largely dependent upon control over heat and mass transfer to generate a controlled temperature profile and controlled working fluid supersaturation profile. The maximum supersaturation achieved governs the "cut-size" for particles (which is generally governed by the Kelvin equation) [17]. The cut-size is often experimentally defined as the size for which the growth and detection efficiency is 0.5; particles larger than the cut-size will efficiently grow and can be detected, and those smaller than it are not detected efficiently. While earlier "cloud chamber" experiments utilized a gas expansion to create supersaturation (which is still used in laboratory measurements [22]), the vast majority of commercially available CPCs generate supersaturation via two distinct methods: (1) turbulent mixing of hot and cold gas streams [23], and (2) controlled heating and cooling of a laminar flow [17,24]. The former method, which was developed in Russia and subsequently Japan several decades ago [25–27], has enabled detection of particles smaller than 2.0 nm in diameter [28–30]. However, turbulent mixing CPCs often experience day-to-day variability in their saturation profiles and cut-sizes, and often have a non-negligible amount of homogeneous nucleation occur during measurement. For this reason, the vast majority of CPCs in use today rely upon heating and cooling of a laminar flow (so-called laminar flow CPCs). In laminar flow CPCs, a nanoparticle laden aerosol is sampled into a tube whose walls are saturated with the working fluid vapor and held at a fixed temperature (the saturator). Subsequently, the particle and vapor laden flow passes into a second tube (the condenser) whose walls are held at a different fixed temperature and also saturated with working fluid vapor. The working fluid typically has a larger molecular weight than that the carrier gas, and the condenser temperature is below the saturator temperature [17,31]. Therefore, heat transfer from the incoming hot, saturated flow to the walls of the condenser occurs faster than mass transfer, leading to supersaturation near the centerline of the flow.

Control over the cut-size is largely application specific (i.e. the goal in CPC design is not always to minimize the cut-size), and since the introduction of laminar flow CPCs [24,31], to control the cut-size while avoiding homogeneous nucleation (which occurs when the supersaturation level is too high), a number of researchers have made modifications and improvements to condensation based detection systems. Notably, Stolzenburg and McMurry [17] introduced a concentric tube geometry and sheath gas to increase the supersaturation and to maximize particle exposure to the supersaturated region. In doing so, they shifted the CPC cut-size with 1-butanol as the working fluid from 10 nm to 2.5 nm. Hering and coworkers developed water based CPCs with similar capabilities to the butanol CPCs [32,33]. Additionally, research focusing on modulation of the working fluid [34], and CPC temperature profiles [19,21] has shown that the cut- size can be pushed below 2.0 nm.

Ultimately, efforts to control the cut-size via changing the working fluid amount to modulation of the Lewis number (Le), which is specifically defined as the ratio of the thermal diffusivity $\left(\frac{k}{\rho C_p}\right)$ to the diffusion coefficient of the working vapor in the gas utilized (D), and typically governs differential heat and mass transfer in vapor laden systems [35–37]. However, prior work has only investigated Lewis number modulation through working fluid modification (or more indirectly through temperature and flowrate changes). A more direct method of probing how differential heat transfer and mass transfer influence the cut-size (and more specifically the activation efficiency curve in a CPC, defined as the fraction of particles grown and detected, as a function of particle size) would be performed by varying the operating gas composition, e.g. by examining performance in variable mixtures of helium and molecular nitrogen. Varying the gas composition dramatically influences the thermal diffusivity and has modest influences on the mass diffusivity of the working fluid. Along these lines, here we examine the influence of Lewis number modulation on heat and mass transfer within a laminar flow CPC operating with 1-butanol as the working fluid. Specifically, we report on measurements of the activation efficiency curve using size selected