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Thermophoretic deposition of airborne particles in an annular flow: experiments and simulations [1]

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Abstract

The thermophoretic deposition of a polydisperse glass aerosol influenced by a fixed temperature gradient in the flow direction was investigated in a prototype thermal cell. The cell was constructed as an annulus and the flow was between the cylinders. The inner cylinder is a high resistance wire electrically connected with a power supply. A fixed thermal gradient between the cylinders can be applied with the outer cylinder maintained at a low temperature and the inner cylinder electrically heated to a higher one. Two optical counters were connected on-line with the cell to perform the measurements. The results were consistent for repeated measurements when the aerosol source is stable and are presented here with an assessment of thermal instability in the horizontally aligned thermal cell.

INTRODUCTION

Thermophoretic deposition occurs when aerosol particles, driven by a temperature gradient in the surrounding gas, move from hot areas onto cold surfaces. The blackening of the glass globe of a kerosene lantern is a common example of the result of this deposition. In industrial applications, it is responsible for the reduced heat transfer efficiency of a heat exchanger, for the low production yield in an aerosol reactor, and for particle loss in a hot aerosol source. It is the working principle in fabricating optical waveguides in the chemical vapor deposition (CVD) process and in sampling with a thermal precipitator.

The phenomenon of thermophoresis was first observed by Tyndall in 1870 as a dust-free air streams arising from a hot body in a chamber containing atmospheric dusts. Lord Rayleigh (1882) showed that this effect

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is due neither to evaporation nor to convection [2]. Finally, Aitken (1884) showed that it is as a consequence of repulsion of particles by a greater molecular bombardment from molecules at a higher temperature [3]. He named this new phenomenon, which occurs in regions of unequal temperature, "thermal repulsion" [4].

The derivation of the theory of thermorphoresis is divided into three dynamic regimes based on the Knudsen number Kn, which is defined as the ratio of the gas mean free path to the particle radius. When the particle size is small in comparison with the gas mean free path ($Kn \rightarrow \infty$), the gas kinetic theory can be applied to calculate the net momentum received by the particle from the gas molecules. The theory of thermophoresis for the free-molecule regime was derived by Waldmann [5] with a momentum transfer method, and was verified experimentally by Schmitt [6] with a modified Millikan cell method. In this case the force depends only on the gas composition.

For particles that are large relative to the gas mean free path ($Kn \rightarrow 0$), a continuum or slip-flow regime, the kinetic theory approach is no longer valid because the presence of the large particles alters the velocity distribution of the gas molecules impinging on the particles. Epstein [7] presented a hydrodynamic approach to the problem by solving the temperature distributions inside and outside a particle. The velocity profile of the gas adjacent to the particle surface was solved using Maxwell's theory of thermal slip as a boundary condition [8]. In their experiments, Schadt and Cadle [9] later showed that Epstein's theory holds for particles of low thermal conductivity ($k_p/k_g < 10$, where k_p and k_g are the thermal conductivities of the particle and the gas, respectively), but not for particles of high thermal conductivity ($k_p/k_g \approx 100$). Note that in the continuum regime the effect depends on both the gas and particle compositions.

To accommodate the differences between experiments and theory for high thermal conductivity particles, four types of analysis have been attempted [10]. The two most famous groups of researchers who derived the theory as well as conducted experiments were the Brock group and the Derjaguin group. Both groups carried out experiments with large particles of high thermal conductivity, but different experimental techniques were used. Brock and Jacobsen [11] used a modified Millikan cell method. Derjaguin et al. [12] suggested a new method, the jet or stream method. Both groups demonstrated agreement between their own experimental results and theory. However, there is a discrepancy between the two sets of results which remains unsettled.

Experimental techniques used in the past can be classified into three types (Fig. 1). The first is the modified Millikan cell method developed by Rosenblatt and LaMer [13]. The famous oil drop Millikan cell was modified to include a thermal gradient to the field of measurement. From this experiment, the thermophoretic velocity of a single airborne particle can

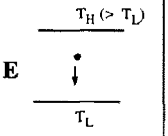
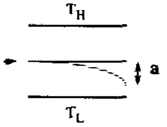
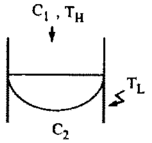
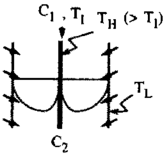
THERMOPHORETIC VELOCITY		THERMOPHORETIC DEPOSITION	
(A). MODIFIED MILLIKAN CELL METHOD	(B). JET METHOD	(C). PENETRATION METHOD	
		LAMINAR	ANNULAR
 ROSENBLATT AND LAMER (1946)	 DERJAGUIN et al. (1966) PRODI et al. (1979)	 ↑ MONTASSIER et al. (1990) ↑ STRATMANN FISSAN (1989) ↓ LIPATOV et al. (1991)	 CHANG et al. (1991)

Fig. 1. Experimental techniques used in thermophoresis studies.

be measured. The second type is the "jet" method invented by Derjaguin et al. [12]. In this method, a thin aerosol stream is injected isokinetically at the exact center of a horizontal, rectangular channel constructed by two parallel plates. The solid line in Fig. 1 is the trajectory in the absence of a thermal field and the dotted line is the trajectory in the presence of a thermal gradient. The thermophoretic velocity is calculated as a function of the vertical displacement of the aerosol stream.

The third type is the deposition of particles in flow along a circular tube. The first experimental measurement to quantify thermophoretic deposition of aerosol in a tube flow was by Byers and Calbert [14], who studied polydisperse dust aerosols in hot turbulent streams. Nishio et al. [15] studied the deposition in flows ranging from laminar to turbulent. In a more carefully controlled experiment, Stratmann and Fissan [16] measured the deposition using monodisperse aerosols in laminar tube flow. The most recent known experimental measurement was by Montassier et al. [17] using a monodisperse fluorescent liquid aerosol, also in a laminar tube flow. In these studies, thermophoretic deposition was influenced by the descending radial temperature gradient in the flow direction. Particle deposition at different locations in the tube, or the overall penetration through the tube, was measured.

In the approach described here, the objective was to quanitfy experimentally the thermophoretic deposition with a fixed radial thermal gradient in an annular flow and to analyze the results with numerical simulations. The variables measured were the aerosol concentrations before and after a prototype thermal cell. The flow was through an annulus so that a fixed radial temperature gradient could be imposed along the flow direction. This provides the major advantage compared to the previous studies in that the gradient is fixed and unambiguous: a numerical simulation is not necessary to determine the thermal gradient.

SIMULATIONS

In all dynamic systems, the thermal velocity, rather than the thermal force, is the key variable. This means that numerical simulation is needed to determine the particle loss. This requires the solution of two simultaneous equations, the energy balance and the general dynamic equations. An implicit finite difference method was used to approximate these equations for numerical simulation. These simulations are discussed in detail in ref. 18. It should be noted that if there is no fixed gradient, additional equations must be solved. Also, for particle size smaller than 0.01 μm diffusional losses are significant.

EXPERIMENTAL DESIGN

The experiment incorporates the following principal components: an aerosol generation/cooling system, a thermal cell system, measuring instruments, and a data acquisition system. A schematic of the entire experimental system is shown in Fig. 2.

The test aerosol is generated from the aerosol generation/cooling system which includes a nebulizer, a furnace, and then a diluter. Intially, a liquid spray of 2% or 8% borosilicate solution is produced from a Collison-type nebulizer. The spary is carried upward, diluted by a filtered nitrogen gas, into a vertical glass column, and is then carried by the flow into a high temperature furnace for evaporation. The droplet evaporates in the

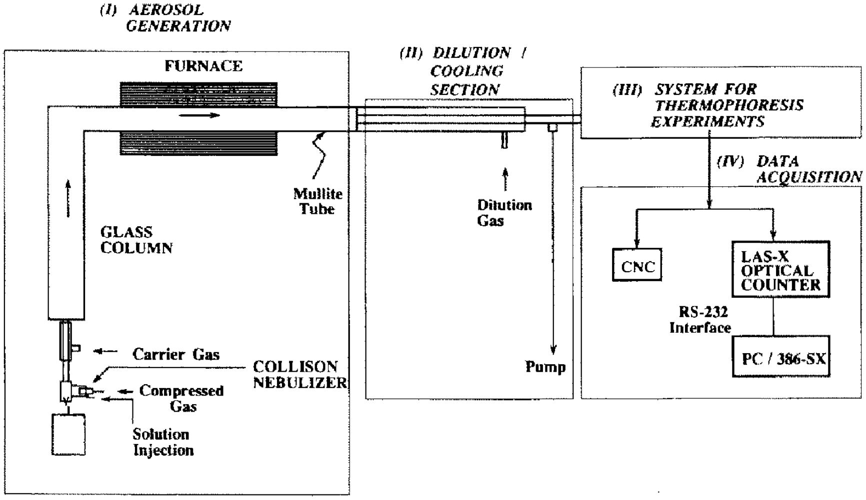


Fig. 2. The entire experimental system.

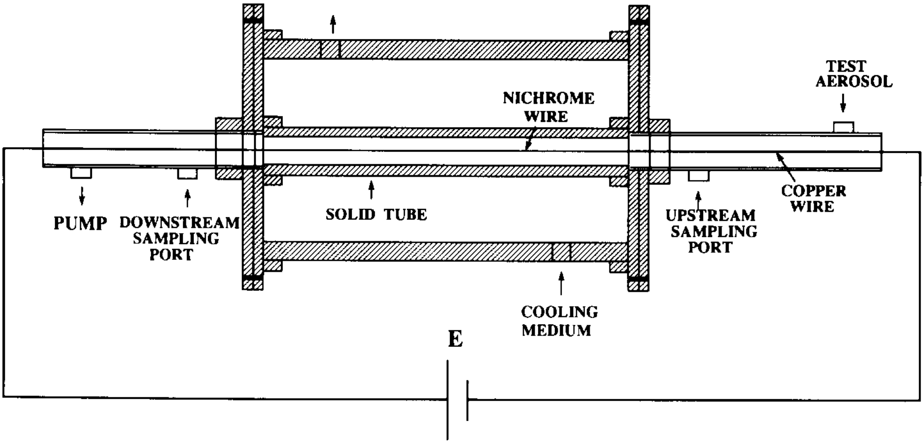


Fig. 3. Thermal cell.

furnace and may shatter if the Rayleigh stability limit is exceeded. The aerosol leaving the furnace is hot, too hot for the instruments and it must be passed through an aerosol diluter for cooling. Cooling is necessary to prevent a significant thermophoretic loss of the test aerosol in the transport line before the thermal cell, as well as for protection of the instruments. Dilution is useful in that it reduces the probability of coincidence error in the optical instruments that determine the particle concentration.

The measurements of thermophoretic deposition are performed in a thermal cell. Two principal aims were considered in the design of the cell: the study of thermophoretic deposition in the cell, with a solid outer tube, and the study of the application of an inward transpiration flow as a means of reducing the thermophoretic deposition and diffusional deposition, with a porous tube. Only the first will be discussed here. The cell was designed and constructed as an annulus equipped with a cooling jacket as shown in Fig. 3. The outer cylinder is either a solid tube or a porous tube depending on the aim of the investigation. It has an inside diameter of 1.2 cm and a total length of 15 cm. The inner cylinder is a high-resistance nichrome wire (Ni:Cr 80:20), 0.0127 cm in diameter (36 BGS; electrical resistance, 86.58 ohm m^{-1}), with the same length as the outer cylinder. Some important features achieved in the design of the cell are: the possibility of replacing the solid outer cylinder with a porous one; the capability of centering the wire in the outer cylinder; the electrical insulation of the inner cylinder, i.e. the bare nichrome wire, from the outer cylinder; and the capability of controlling the inner cylinder temperature in situ with a Wheatstone bridge. Because the wire expands on heating the wire must be spring loaded to hold it taut.

The thermal gradient was determined from the measurement of the temperatures at both the outer and inner cylinders. The outer cylinder was maintained at the outlet temperature of the circulating water because the difference between the temperatures of the cooling water measured at the inlet and outlet of the cooling jacket was negligible. The temperature of the inner cylinder was controlled and measured using a Wheatstone bridge.

The deposition measurements were conducted with two on-line instruments, a continuous condensation nuclei counter (CNC, TSI 3020) and a laser aerosol spectrometer (LAS-X, PMS). Data acquisition with the LAS-X counter was available via its built-in RS232 to a personal computer. The quantities measured were aerosol concentrations before and after the thermal cell. The continuous measurements of the concentrations at two different locations were possible using a 4-way ball valve to switch between the sampling sites.

EXPERIMENTAL MEASUREMENTS

The principal goal is to measure the deposition due solely to thermophoresis. To ensure this, four types of measurements are made sequentially, as follows (1) A measurement to ensure that there is no other aerosol source in the system other than the glass aerosol, after the test aerosol is generated. (2) A measurement to check the stability of the test aerosol concentration after the aerosol source becomes stable. (A stable aerosol source is crucial to obtain the true magnitude of the thermophoretic deposition.) (3) A measurement to obtain the deposition with no temperature gradient applied in the cell; and finally (4) a measurement of the deposition with a fixed temperature gradient. The thermophoretic deposition can be determined from the last two measurements.

There is a possible onset of thermal instability because the fluid above the hot wire is cooler than that at the wire surface. The density near the wire is therefore lower, and there is a buoyancy effect. The Rayleigh number *Ra* is the criterion for estimation of the onset of thermal instability [19]. Results reported by Lipatov et al. [20] indicate that this instability is a real possibility. Numerical simulations were carried out to test this possibility.

RESULTS AND DISCUSSION

Typical experimental results were obtained with a solid glass aerosol generated at furnace temperature between 500 and 600°C. The deposition efficiency $(1 - C_2/C_1)$ was determined from the ratio of the aerosol concentrations before C_1 and after C_2 the cell. The aerosol concentration measurements (before and after the cell) were repeated continuously. The total sampling time for measurement at each sampling site was one minute.

the possible onset of thermal instability was numerically simulated for both cases. The maximum local Rayleigh number in the two cases discussed here was found to be about 1050 which is well below the critical Rayleigh number of 1717, and occurs at the outer region above the hot wire.

In Fig. 4 the total deposition as determined with the CNC counter is plotted as a function of sequential measurement. In this experiment, the Reynolds number *Re* of the gas flow in the cell is about 157, and the residence time is about 1 s. The data obtained from the CNC counter is the total concentration of nuclei of sizes down to 0.01 μm. In this case, the aerosol source was fairly stable. The total deposition for twenty sequential measurements was around 18%–21%.

In Fig. 5 the fractional deposition as determined with the LAS-X counter is plotted as a function of particle size, in this case *Re* = 76 and *t* = 2 s. The data obtained from the LAS-X counter are particle counts in sixteen discrete size intervals from 0.09 to 3 μm. The result showed that the fractional deposition efficiency for the four sizes illustrated here ranged from 18% to 25%. For particle sizes of less than 0.25 μm, particle counts were unstable in most measurements so that the true effect of thermophoresis cannot be evaluated. For particles larger than 0.6 μm, negative efficiencies were obtained regardless of whether a thermal gradient was imposed. This may be due to a virtual impaction effect at the sampling site before the cell, where the sampling port is at a 90° angle facing the mainstream.

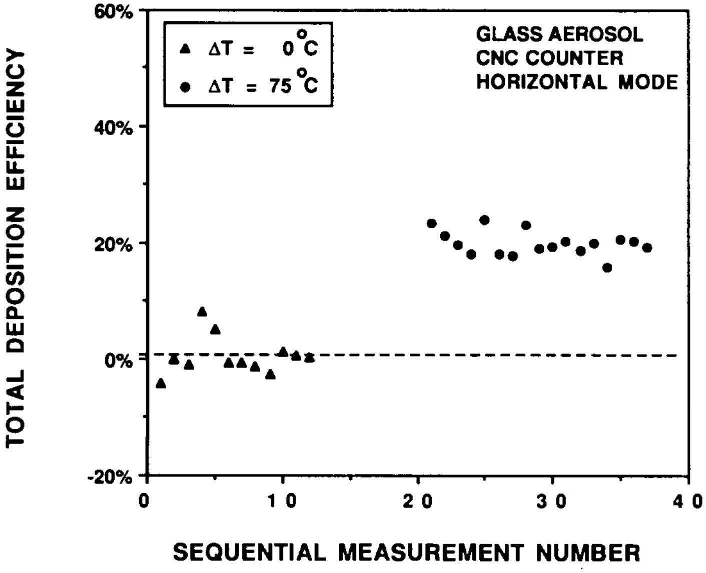


Fig. 4. Total deposition efficiency measured sequentially, determined with a CNC counter for temperature differences between the cylinders (ΔT) of 0°C and 75°C.

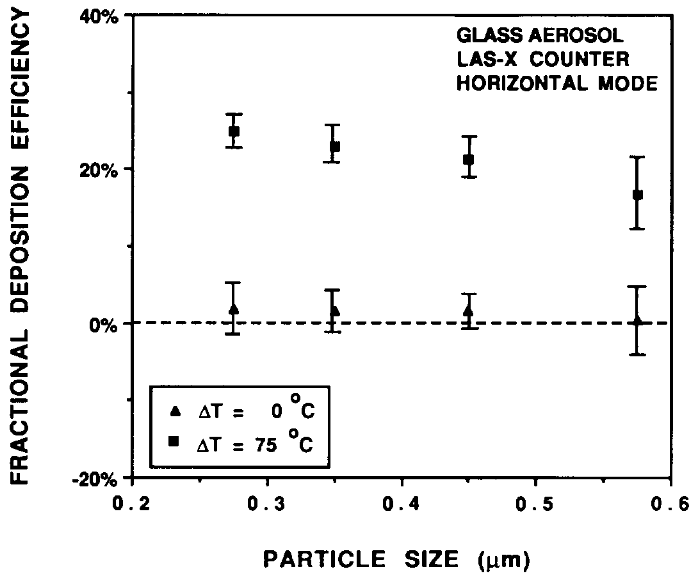


Fig. 5. Fractional deposition efficiency as a function of particle size, determined with a LAS-X counter for temperature differences between the cylinders (ΔT) of 0°C and 75°C.

SUMMARY

A prototype annular thermal cell which incorporates a fixed thermal gradient was constructed. It was used successfully in the quantitative study of the thermophoretic deposition of a polydisperse aerosol under the influence of a fixed thermal gradient in the flow direction. An experimental procedure was developed to determine the deposition due solely to thermophoresis. The experimental system allows on-line, continuous measurement of aerosol penetration through the thermal cell and, therefore, repeated measurements of thermophoretic deposition for a fixed thermal gradient. Consistent results of both the total deposition efficiency as determined with a condensation nucleus counter, and the fractional deposition efficiency as determined with a laser aerosol spectrometer were obtained for repeated measurements when the fluctuation in the aerosol source was insignificant in comparison to the magnitude of the deposition owing to thermophoresis. A numerical analysis to examine the possibility for the onset of thermal instability in the cell was performed for the experiments reported here. It was found that the maximum possible Rayleigh number for both cases was well below the critical Rayleigh numbers in Bernard instability and couette flows.

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