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The Measurements of Thermophoretic Deposition of Aerosol Particles in the Continuum and Slip Flow Regions

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Abstract. The thermophoretic deposition of limestone particles onto a cold plate above which a temperature gradient was established has been measured. The experimental setup and methodology are described. The particle size distribution was measured by means of an Andersen impactor. The concentration of particles of different sizes was also measured. The Knudsen numbers (Kn) covered both continuum and slip flow regimes ($Kn = 1 \times 10^{-2}$ to 0.93). The theoretical expressions for thermal velocity of particles in the two regimes were calculated using the theories of Derjaguin and Yalamow and Brock, respectively. The measured concentration of particles of each size (0.4 - 9 μm) along with theoretical values of thermal velocity were used to predict the total particle flux. The comparison between the directly measured particle flux and the predicted values showed good agreement. This result validates the thermophoresis theoretical expressions which have been proposed for both flow regimes involved in the present investigation.

Nomenclature

C = slip correction factor
C_m = coefficient of viscous slip
C_t = temperature jump coefficient
d_p = particle diameter
F = frictional resistance of the gas
F_{th} = thermal force
K = translational part of the thermal conductivity
K_g = thermal conductivity of gas
Kn = Knudsen number = λ/r_0
K_p = thermal conductivity of particle
M = molecular weight of gas
P = pressure

r_p = particle radius
R = universal gas constant
T = absolute temperature
V_g = gas velocity
V_t = thermal velocity
y = normal distance measured from the surface

Greek letters

λ = mean free path
μ = dynamic viscosity of gas
π = 3.1415
ρ = gas density
ρ_p = particle density

1. Introduction

Aerosol particles experience thermal forces when they are suspended in a gas with a thermal gradient. This phenomenon-thermophoresis – has been a subject of many investigations. It has also been applied in the development of aerosol samples.

The description of the thermal force is usually determined by the dimensionless parameter called Knudsen number (λ/r_p). Many investigators offered different theoretical expressions to describe quantitatively the thermal force on aerosol particles [1-6]. The range of applicability of each expression has been given in terms of Knudsen number as follows: in the free molecular regime ($\lambda/r_p >> 1$); in the continuum regime ($\lambda/r_p << 1$); in the slip flow regime ($\lambda/r_p < 1$), and in the transition regime ($\lambda/r_p \simeq 1$). However, it has been claimed that the thermal force is not effective for the precipitation of particles larger than about 10μm [7], and later El-Shobokshy [8] showed experimentally that aerosol particles larger than about 8 μm are not affected by the thermal force.

The thermophoretic velocities of aerosol particles in the transition region have been measured by Prodi *et al.* [9]. Their results agreed with Brock's theory for the particle diameter range 0.20 - 1.0 μm and the associated Knudsen number range was 0.13 - 0.64 according to the experimental conditions.

In many real situations aerosol particles with a wider size range are usually present, consequently different regimes are involved. In such a case the thermophoretic velocities should be predicted by the applicable expression in each regime.

It is therefore, worthwhile to measure the thermophoretic deposition of aerosol particles under experimental conditions in which more than one regime are applicable. Thermophoretic velocity of particles in each regime can be determined by the corresponding theoretical expressions, in order to examine the validity of each and also the possibility of estimating the particle flux under the combined regimes.

2. Theoretical Model Review

The theories of thermophoresis consider separately two limiting cases of spherical aerosol particles which are either small or large in comparison with the mean free path of the gas. For the intermediate case ($r_p = \lambda$) a theory has also been developed [1].

For small particles the theory is indeed simple and complete, however, for large particles and the intermediate case, the theories still contain uncertainties.

Table 1 summarizes the available theories of thermophoresis and the expressions for the thermal force along with the conditions for which they are applicable.

Figure 1 gives a quantitative comparison of the thermal velocity per unit thermal gradient calculated by the various theoretical equations.

The thermal velocity of a particle (V_t) is determined by equating the thermal force (Table 1) and the frictional resistance of the gas (Knudsen-Weber equation):

$$F = 6\pi\mu r_p V_t \; [1 + 2.5\,(\frac{\lambda}{d_p}) + 0.84\,(\frac{\lambda}{d_p}) \exp(-\; 1.74\,(\frac{\lambda}{d_p}))]^{-1}$$

The effect of discontinuity in the tangential velocity of molecules at the surface of particles has to be accounted for by introducing a slip correction factor, C. The empirical correlation of Davis [10] which is based on the weighted average of experimental falling speeds may be used for this purpose, this is:

$$C = 1 + \frac{2\lambda}{dp}\{1.257 + 0.4 \exp\;(-1.1\frac{d_p}{2\lambda})\}$$

The mean free path length of the gas λ is calculated using the following equation

$$\lambda = \frac{\mu}{0.499\,p}\,(\frac{\pi M}{8\,R\,T})^{1/2}$$

3. Experimental Method and Procedure

Figure 2 shows schematically the test apparatus used in the investigation. The experimental hot and cold surfaces are separated by a gap of 3 mm. The hot plate is heated by an evenly distributed set of electrical heaters fixed at the opposite face of the plate and is well insulated. The plate is provided with three thermocouples fixed

Table 1. Summary of thermophoresis theoretical expressions

Thermal force on aerosol particles	Range of applicability	Investigator
$F_{th} = -\frac{1}{2}(\pi r_p^2)\ (P\lambda/T)\ \frac{dT}{dy}$	$\frac{\lambda}{r_p} >> 1$ (Free molecular regime)	Cawood (1936)
$F_{th} = \frac{-32\, r_p^2\ k}{15\, V_g}\frac{dT}{dy} = -\ 4 r_p^2\ (\frac{P\lambda}{T})\frac{dT}{dy}$	$\frac{\lambda}{r_p} >> 1$ (Free molecular regime)	Waldmann (1959) Derjaguin & Bakanov (1962)
$F_{th} = -(9\pi)\ (r_p)\ (\mu^2/\rho T)\ (\frac{K_g}{2K_g + K_p})\frac{dT}{dy}$	$\frac{\lambda}{r_p} << 1$ (Continuum regime)	Epstein (1929)
$F_{th} = -(\frac{3\pi\mu^2 r_p}{\rho T})\ [\frac{8K_g + K_p + 2C_t(\frac{\lambda}{r_p})K_p}{2K_g + K_p + 2C_t(\frac{\lambda}{r_p})K_p}]\frac{dT}{dy}$	$\frac{\lambda}{r_p} << 1$ (Continuum regime)	Derjaguin & Yalamov (1965)
$F_{th} = \frac{(-12\pi)(\mu r_p^2)(K_g/K_p + 2C_t\lambda/r_p)(C_m\lambda/r_p)}{(1 + 3C_m\lambda/r_p)(1 + 2K_g/K_p + 2C_t\lambda/r_p)}\ \frac{dT}{dy}$	$\frac{\lambda}{r_p} < 1$ (Slip flow regime)	Brock (1962b)
$F_{th} = -9\pi\mu r_p^2(\frac{2R}{\pi T})^{1/2}\frac{\lambda}{r_p}\ \frac{1}{1 + 3C_m(\frac{\lambda}{r_p})}\ \frac{(K_g/K_p) - C_t(\lambda/r_p)}{1 + 2(\frac{K_g}{K_p}) + 2C_t(\frac{\lambda}{r_p})}$	$\frac{\lambda}{r_p} \approx 1$ (Transition regime)	Brock (1967)

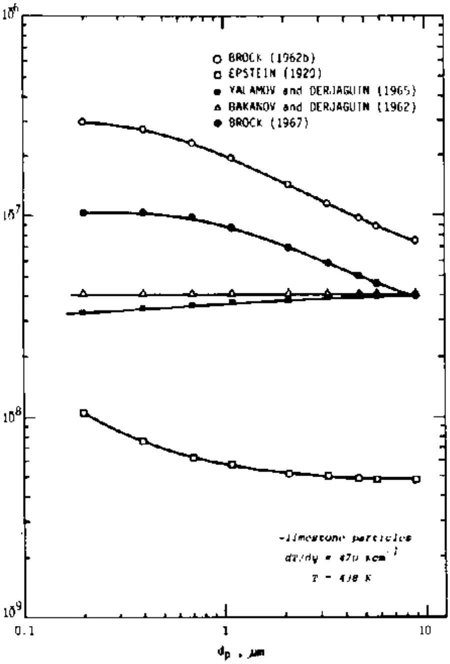

Fig. 1. Thermal velocity per unit thermal gradient for different theories

at a depth of 2 mm from the surface for measuring the temperature. The cold plate is cooled by an open circuit tap water flow at a rate of 8 *l* min^{-1} where the water jacket volume is only 0.0034 m^3. This plate is provided also with three thermocouples for temperature measurement. A thin aluminium foil was used as a deposition surface. A piece of foil 40 × 40 mm was fixed firmly to the cold plate by means of a frame which uncovers only this area of the foil. The electric power supplied to the hot plate was adjusted to the required level by means of a regulator. All thermocouples were connected to a multi-channel chart recorder. The recorder provides a digital printout of temperatures in °C every one minute during the test period. The test apparatus was located in an isolated space in the laboratory in which a fine dust of limestone prepared specially for this investigation was uniformly dispersed throughout the space. A fan was used to supply the dust to the test space, stir the air and maintain the uniformity of dust dispersion. An 8-stage Andersen cascade impactor was used for particulate sampling. The sampler's inlet tube was connected to the lower end of the gap separating the hot and cold plates, this assures that the particulate concentration measured represents the average concentration within the gap. The impactor

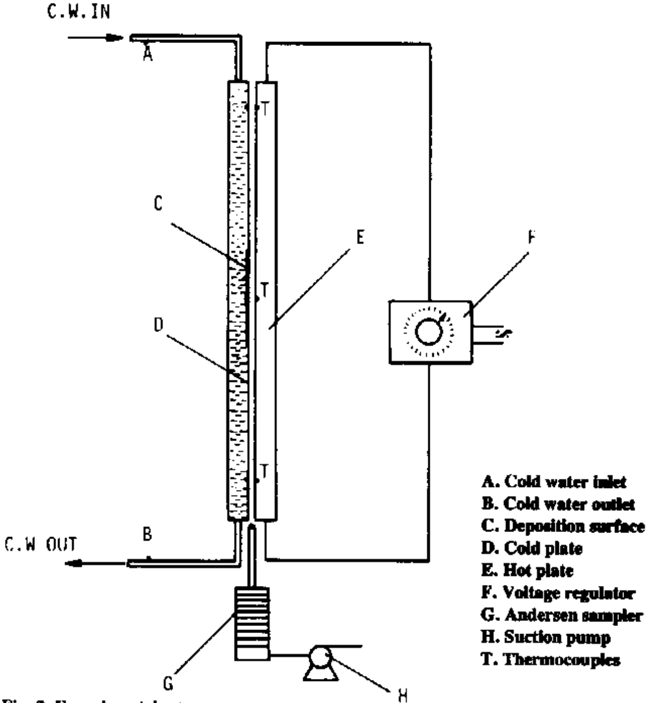

Fig. 2. Experimental setup

operates at a constant flow rate of 28.3 l.min^{-1} (1 ft^3 .min^{-1}) with calibrated effective cut diameters (ECD) of 9, 5.8, 4.7, 3.3, 2.1, 1.1, 0.7, and 0.4 μm. An 81 mm fiber glass filter was used as a back-up filter for collecting particles smaller than 0.4 μm. The impaction surfaces used in the impactor were Aluminium foils covered with very thin layers (finger print) of vaseline. Several sets of impaction surfaces and filters were prepared, dried and weighed before the test. The test was started by first supplying the desired electric current to the hot plate and circulating the cooling water. The chart recorder print out was observed until a thermal equilibrium prevails before starting the test run. The Andersen sampler is then loaded by impaction surfaces, a filter located in place and the dust is dispersed. The sampler is then operated for about 2 hours after which the impaction surfaces, the filter and the deposition surface are dried before reweighing them to determine the mass of collected particulates on each. The average temperature of both hot and cold plates during the test is determined from the recorder print out. A computer program was developed to process the experimental data and to produce the following results:

a) The particulate size distribution and its related parameters.
b) Mass concentration of each particle size (based on aerodynamic diameter).
c) Experimental particulate flux on the deposition surface in $\mu gm^{-2} s^{-1}$.
d) Thermophoretic velocity of the particles calculated by the various theoretical equations, and
e) The total theoretical flux.

For estimating the individual flux of particles of different sizes, the proper theoretical expression has been selected by the computer based upon the value of Knudsen number, from which the total particle flux was computed. The air properties were determined within the computer program as a function of the experimental temperature. The thermal conductivity of limestone particulate was assumed to be the same as that of the bulk of the material. Some visual tests under an optical microscope showed that most dust samples contain spherical particles.

4. Results and Discussion

The particulate data for the Andersen impactor have been analysed and plotted on a probability graph as shown in Fig. 3. It has to be noted that the particle size distributions were almost identical for each test run and Fig. 3 shows a typical size

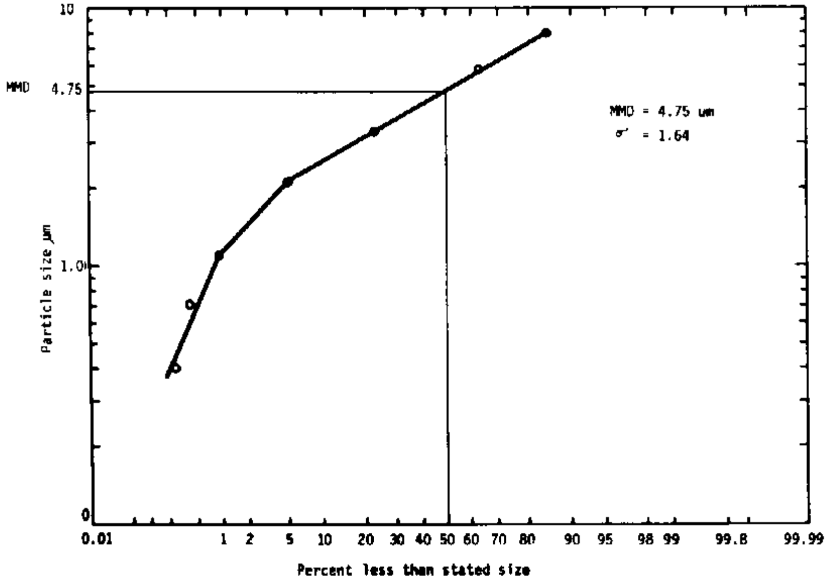

Fig. 3. Typical limestone particle size distribution

analysis for the limestone aerosol used. The mass median diameter of the aerosol is 4.75 μm with a geometric standard deviation of 1.64. The concentration of particles of different sizes in the eight stages of the impactor as well as those collected on the backup filters were also determined in each test run.

The thermal velocity of particles was calculated using the appropriate theoretical expression depending on the value of Kn (λ/r_p). In the present experiment, Kn was in the range of 1×10^{-2} to 0.93, for which both continuum regime and slip flow regime are considered to be present.

The flux of particles of each size was then calculated by multiplying the calculated particle's thermal velocity by the measured concentration. The total particle flux was determined by summing up all the individual flux, and this is considered as a theoretical value since it has been calculated using theoretical thermal velocities.

The experimental total (including all sizes) particle flux was determined as the difference betwen the weight of the deposition surface after and before the experiment divided by the (deposition area × exposure time). A Mettler microbalance having a minimum measureable weight of 1 μg was used for this purpose. The theoretical and experimental particle flux values are plotted against each other in Fig. 4. The thermal gradient was kept nearly constant at a value of 470 Km^{-1}.

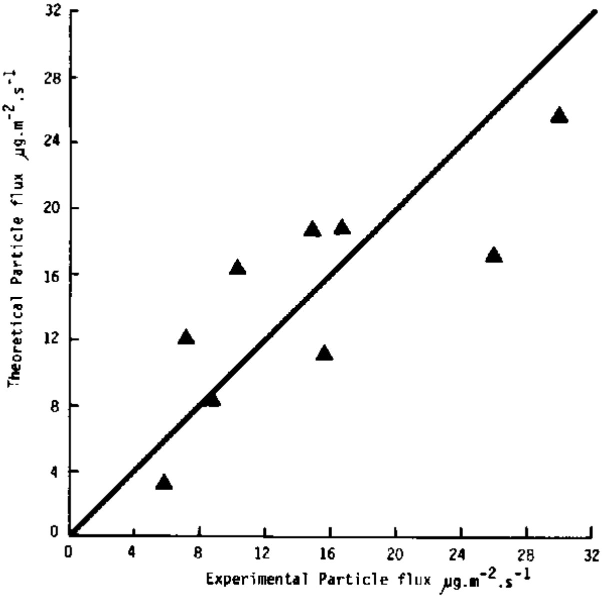

Fig. 4. Comparison between theoretical and experimental particle flux

Figure 4 shows a reasonable good agreement between the experimental and theoretical values of particle flux. Such results validate both theoretical expressions for the particle thermal velocity by Derjaguin and Yalamov (1965) for the continuum regime ($\lambda/r_p << 1$) and that of Brock (1967) for the slip flow regime ($\lambda/r_p < 1$).

For perfect agreement all experimental and theoretical points should lie on the solid line, the deviations, however, have occured due to some interferences which are believed to be present.

a) Non-sphericity of particles; all thermophoretic theories assume spherical particles, a matter which by any means cannot be guaranteed when the limestone aerosol were produced during the crushing, grinding and drying processes.

b) Inaccurate representation of the physical properties of particles material (density and thermal conductivity) which include some impurities.

c) The agglomeration of particles, a phenomenon which more likely takes place as the particles flow through the experimental channel.

d) Particles may carry electric charges, and this of course affect the deposition of particles, a matter which cannot be quantified under the circumstances of the present experimental conditions.

Inspite of all the above interferences the theoretical expressions for the thermal velocity of particles for both continuum and slip flow regimes have successfully estimated the flux of polydispersed particles. The advantage of using the Andersen impactor is that it separates the particles into eight stages, each having a known aerodynamic diameter and mass concentration. This provides a reasonable accuracy when estimating the thermal velocity of particles and their flux.

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دراسة ظاهرة الترسيب الحراري لذرات الغبار الجوي في كل من التيار المتصل والانسيابي

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ملخص البحث: في هذا البحث تمت دراسة وقياس ترسب ذرات الغبار الجيري على سطح بارد بعد إيجاد تدرج حراري عليه. وتم شرح الجهاز المستخدم ونظرية البحث، وقد تم قياس التوزيع الحجمي للذرات على حسب قطرها باستخدام جهاز تصنيف الذرات المسمى (اندرسون امباكتور) وتم أيضا قياس تركيز الذرات ذات الأحجام المختلفة.

وقام الباحثان بعمل التجارب لأرقام (اندرسون) مابين ١×١٠^{-٢} الى ٩٣ر٠ لتغطي كلا من التيار المتصل والانسيابي. كما تم دراسة المعادلات النظرية وخاصة نظريات (ديرجاجون ـ ويالاموف وبروك) لحساب السرعة الحرارية وباستخدامها واستخدام تركيز الذرات تم حساب تدفق تلك الذرات.

ولقد وجد أن هناك توافقًا جيدًا بين القيم التي تم قياسها والقيم التي تم حسابها من المعادلات النظرية وهذا يعضد مركز تلك النظريات في كل من التيارين (المتصل والانسيابي).