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Investigation of Thermal Accommodation using Laser-Induced Incandescence

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ABSTRACT

Accurate particle sizing through laser-induced incandescence (LII) requires an understanding of the thermal accommodation coefficient; if the particle size is known a priori, however, LII can instead be used to measure this quantity. This paper presents thermal accommodation coefficient data between soot and different gases, which are used to infer how gas molecular mass and structure affects energy transfer during gas-surface scattering.

INTRODUCTION

The thermal accommodation coefficient, α_T, specifies the energy transferred when an incident gas molecule collides with a surface composed of thermally-vibrating atoms. Knudsen [1] was the first to formally define this parameter as

$$\alpha_T = \frac{T_{g,o} - T_g}{T_s - T_g} = \frac{E_{g,o} - E_{g,i}}{E_s - E_{g,i}}, \tag{1}$$

where T_g and $T_{g,o}$ are the kinetic temperatures of incident and scattered molecular streams, T_s is the kinetic surface temperature, and $E_{g,i}$, $E_{g,o}$, and E_s are the energies corresponding to these temperatures.

Recent interest in thermal accommodation coefficients is largely driven by time-resolved laser-induced incandescence, an emerging technique for sizing aerosol particles. In this procedure, a nanosecond laser pulse energizes aerosol particles to incandescent temperatures, typically 3000-3500 K in low fluence experiments, and the resulting spectral incandescence is measured as the particles cool by conduction with the surrounding gas. Because the cooling rate of a particle is proportional to its specific surface area, the particle size distribution can be inferred directly from the observed incandescence decay rate. To obtain accurate particle size measurements, however, it is crucial to accurately model the heat conduction between the energized particles and the gas, a process that is governed by the thermal accommodation coefficient.

Conversely, laser-induced incandescence can be used to measure α_T if the aerosol particle size is known *a priori* through electron microscopy or other independent means. In this study, time-resolved LII is used to measure α_T values between soot sampled from a well-characterized laminar diffusion flame and various monatomic and polyatomic gases. The effect of gas molecular mass and structure on thermal accommodation are then inferred from trends in the data. This knowledge elucidates the underlying physics of thermal accommodation, which must be understood for LII to mature into a reliable tool for aerosol particle sizing.

EXPERIMENTAL PROCEDURE

The experimental apparatus is shown schematically in Fig. 1. The ethylene laminar diffusion flame is generated with a Gülder burner operating under conditions described in [2]. The sampling system consists of a venturi mini-eductor with a 0.030 inch critical diameter and a soot sampling probe connected to the venturi throat. Motive gas flows through the venturi at 20 litres per minute, causing a pressure drop at the venturi throat that pulls soot from the flame centerline at a height of 52 mm above the burner into the motive gas stream. Although gas from the flame and the surroundings is also entrained into the sampling system, a large dilution ratio ensures that it has a negligible influence on the experimental results.

The motive gas and soot then enter the LII chamber, where the soot particles are energized with a pulsed ND:YAG laser operating 16 mJ per pulse at 20 Hz and 1024 nm. The laser heats the soot to a peak temperature of approximately 3600 K, and the resulting incandescence is imaged onto two photomultipliers equipped with narrow-band interference filters centered at 397 nm (36 nm FWHM) and 782 nm (19 nm FWHM), respectively. Transient signals from the photomultipliers are digitized and averaged over 100 shots to mitigate shot noise resulting in a set of incandescence decay curves and a pyrometrically-defined temperature decay curve.

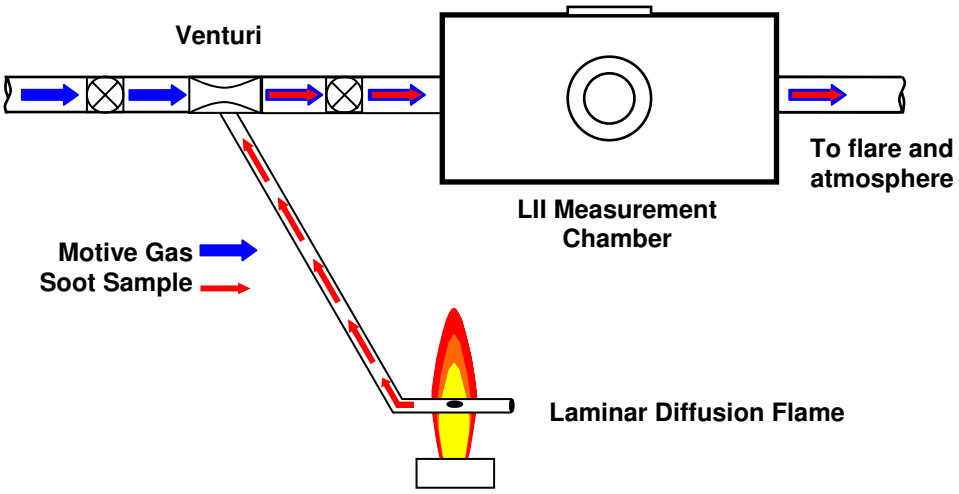

Figure 1: Schematic of experimental apparatus.

The LII data is analyzed following the procedure described in [2]. After the laser pulse, the soot cooling rate is governed by

$$\rho_s c_s N_p \frac{\pi d_p^3}{6} \frac{dT_p}{dt} = -q_{cond}\left[T_p(t) - T_g\right], \tag{2}$$

where ρ_s and c_s are the density and specific heat of soot, N_p is the number of primary particles per soot aggregate, d_p is the primary particle diameter, and q_{cond} is the net heat transfer from the aggregate to the surrounding gas. The density and specific heat of soot are taken to be 1900 kgm^{-3} and 2100 $Jkg^{-1}K^{-1}$, respectively, while N_p and d_p are set equal to 42 and 29 nm based on the TEM study in [3]. Heat conduction in the free molecular regime is given by

$$q_{cond}\left[T_p(t)-T_g\right]=\alpha_T\,A_{cond}\,\frac{P_g\,c(T_g)}{8T_g}\,\frac{\gamma^*+1}{\gamma^*-1}\left[T_p(t)-T_g\right], \tag{3}$$

where $c(T_g) = (8k_BT_g/\pi m_g)^{1/2}$ is the mean speed of gas molecules incident on the particle surface and γ^* is an average adiabatic constant defined in [4]. (Pressure and temperature are assumed to be 101.3 kPa and 300 K.) The effective heat transfer area of the soot aggregate is $A_{cond} = \pi d_p^2(N_p/1.10)^{-1.08}$ [5], which accounts for the greater accessibility of primary particles on the aggregate periphery to incident gas molecules compared to those within the interior.

The thermal accommodation coefficient is found by first setting $\theta(t) = T_p(t) - T_g$ and then solving Eqs. (2) and (3) to get $\ln\theta(t) = -\alpha_T C_1 t + C_2$, where

$$C_1=\frac{3}{4}\,\frac{A_{cond}}{\rho_s\,c_s\,N_p\,\pi\,d_p^3}\,\frac{P_g\,c(T_g)}{T_g}\,\frac{\gamma^*+1}{\gamma^*-1}, \tag{4}$$

and C_2 is a constant of integration. The thermal accommodation coefficient is then found from a linear regression on $\ln\theta(t)$ with respect to cooling time, t, as shown in Fig. 2. This procedure is complicated by the fact that the observed LII signal at any instant is due to the integrated incandescence of different-sized particles cooling at rates proportional to their specific surface area. Provided the laser field is spatially uniform particles of all sizes initially cool at the same exponential rate, but at longer cooling times the pyrometric temperature decay becomes progressively non-exponential due to the non-uniform cooling of different-sized particles. There is also a commonly-observed and as yet unexplained "anomalous" cooling rate lasting 10-25 ns after the laser pulse that is particularly pronounced in LII experiments on aerosols at ambient temperatures. To address both these issues, the linear interpolation is carried out using data sampled from 50-100 ns after the peak soot temperature.

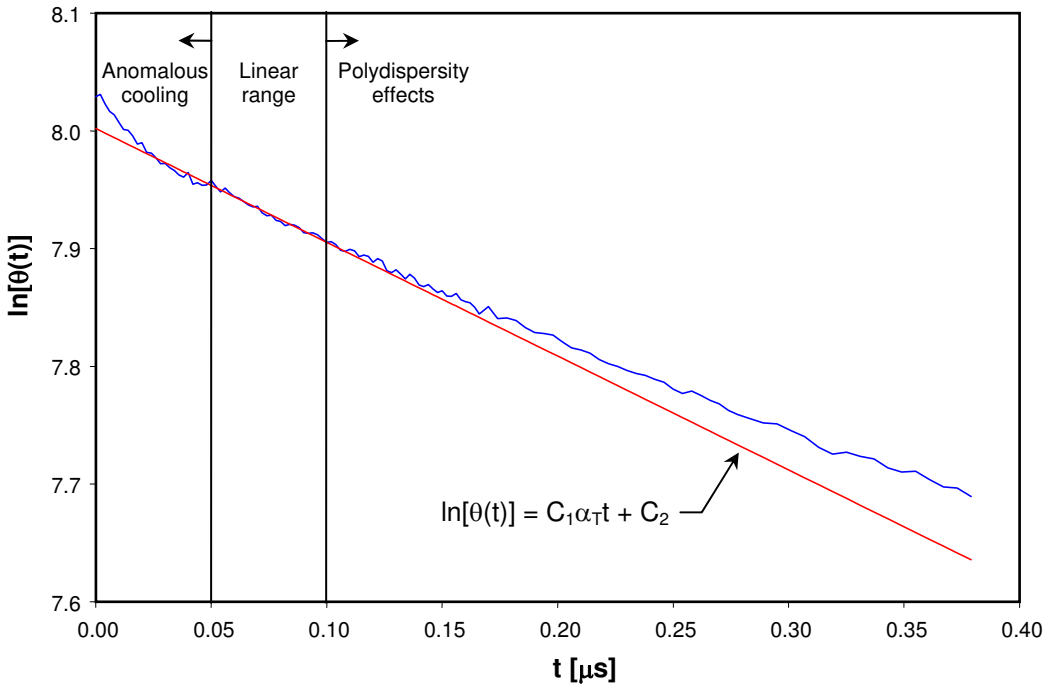

Figure 2: Linear regression of the pyrometrically-derived soot temperature.

DATA ANALYSIS

Figure 3 shows that α_T is a function of the mass and structure of the gas molecule. For monatomic gases, α_T increases monotonically with m_g due to the surface deformation rate as governed by the incident gas molecular speed, $u_{g,i}$, and not the incident molecular momentum or energy as one may intuitively expect. A gas molecule striking the surface with a high incident speed causes rapid surface deformation, requiring the gas atom to do substantial deformation work on the surface that deducts from the energy transferred from the vibrating surface to the gas atom. In a slow collision, on the other hand, the surface is more compliant since the surface atoms can move in phase with the gas molecule and relatively little collision energy is lost to deformation work. Consequently, as m_g increases $u_{g,i}$ decreases resulting in more efficient energy transfer between the vibrating surface atoms and the gas molecule, and a larger value of α_T. This mechanism has also been hypothesized by other researchers [6-8].

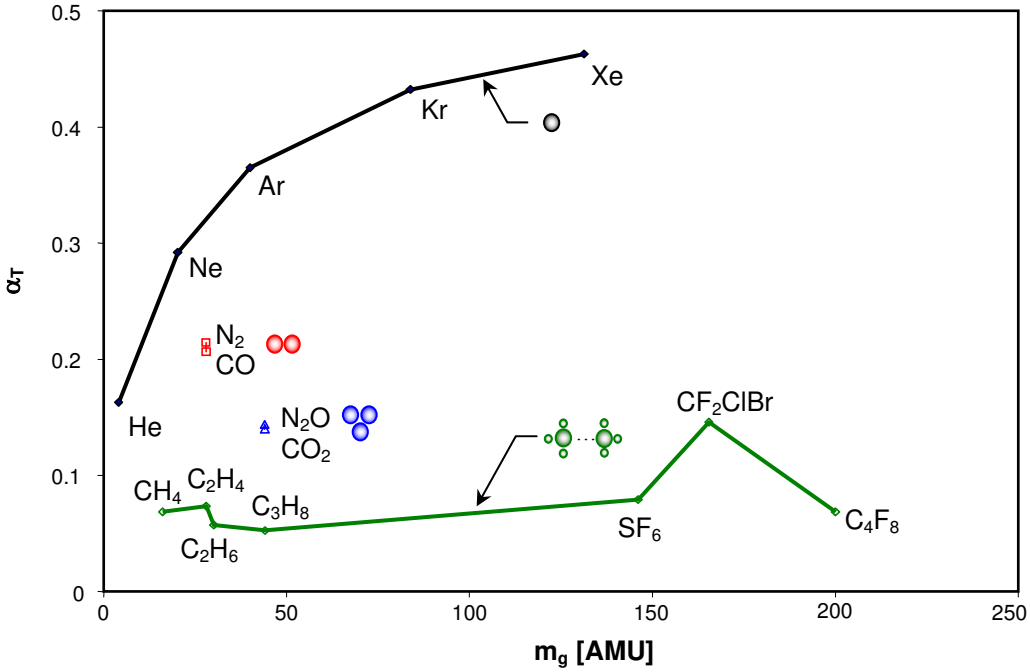

Figure 3: Thermal accommodation coefficients between soot and various gases.

In addition to possessing translational energy, polyatomic gases also have rotational and vibrational energy, collectively referred to as internal energy. Most LII studies assume that energy from the thermally-vibrating surface is transferred to the translational and internal modes of the gas molecules with equal efficiency, but Fig. 3 demonstrates that this is not the case since α_T is lower for more structurally-complex molecules.

Knudsen [9] defined separate thermal accommodation coefficients for translational and internal energy modes,

$$\alpha_{T,trans} = \frac{E_{trans,o} - E_{trans,i}}{E_{trans,s} - E_{trans,i}}, \quad \alpha_{T,int} = \frac{E_{int,o} - E_{int,i}}{E_{int,s} - E_{int,i}}, \tag{6}$$

based on the fact that a Maxwellian molecular steam at kinetic temperature T advects $E_{trans} = 2k_BT$ of translational energy and $E_{int} = 2/3\zeta_{int}(T)k_BT$ of internal energy per molecule. The number of active internal energy modes, $\zeta_{int}(T)$, is obtained from the adiabatic gas constant,

$$\zeta_{int}(T) = \frac{5 - 3\gamma(T)}{\gamma(T) - 1}. \tag{7}$$

(The terms $E_{trans,s}$ and $E_{int,s}$ are the translational and internal energy per molecule carried by a molecular stream at T_s, and represent an approximate limiting case.) According to Eq. (6), the thermal accommodation coefficient for internal energy is found directly from $E_{int,o}$,

$$E_{int,o} = E_o - E_{trans,o} = \alpha_T(E_s - E_i) + E_i - \left[\alpha_{T,trans}\left(E_{trans,s} - E_{trans,i}\right) + E_{trans,i}\right], \tag{8}$$

provided that $\alpha_{T,trans}$ is also known. The translational thermal accommodation coefficient for each polyatomic gas is estimated from the set of α_T values for monatomic gases, based on the assumption that $\alpha_{T,trans}$ depends only on m_g through $u_{g,i}$, and is independent of gas molecular structure. Figure 4 shows that $\alpha_{T,int}$ is substantially lower than $\alpha_{T,trans}$, and furthermore, even though the amount of internal energy transferred to the gas molecule generally increases with the number of available internal degrees of freedom, $\alpha_{T,int}$ tends to decrease with increasing $\zeta_{int}(T_g)$. This shows that energy is preferentially accommodated by the rotational modes over the vibrational modes since the latter represent a larger portion of the active internal energy modes as the molecular structure becomes more complex. This is consistent with the results of molecular beam scattering experiments involving SF_6/graphite [10] and CF_3Br/graphite [11] and is not surprising given the long relaxation times of vibrational energy modes [12, 13], although further molecular dynamics studies are needed to show how the collision mechanics influence vibrational accommodation.

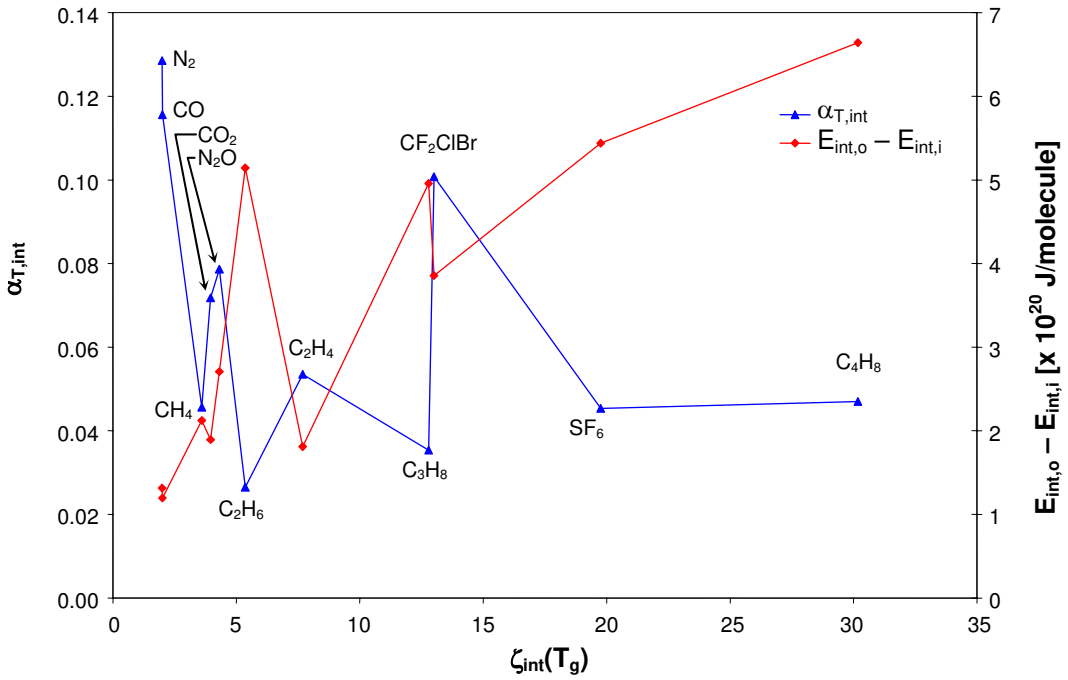

Figure 4: Plot of $\alpha_{T,int}$, and $E_{int,o}$ as a function of internal degrees of freedom, $\zeta_{int}(T_g)$.

CONCLUSIONS

This study presents thermal accommodation coefficients between well-characterized soot and various gases measured using time-resoled laser-induced incandescence. Analysis of experimental data shows that thermal accommodation between soot and monatomic gases increases with increasing molecular mass due to its influence on collision speed and surface deformation rate. The thermal accommodation coefficients between soot and polyatomic gases are lower than the monatomic gas values because energy is transferred from the vibrating surface atoms to the translational energy modes of the gas molecule more readily compared to the rotational and vibrational energy modes. This information will lead to better understanding of the physics behind thermal accommodation coefficients in LII studies. Future work will include expanding the experimental dataset to include measurements of thermal accommodation coefficients at different gas temperatures and developing a molecular dynamics simulation, with the ultimate goal of providing benchmark α_T values for time-resolved LII studies.

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