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Investigation of fine scale structure of turbulent and molecular diffusion in coaxial jets of He/CO_2 in air by LDA and Rayleigh scattering

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ABSTRACT

This paper revisits the important issue of differential diffusion and provides new experimental results and subsequent analysis that attempts to quantify the relationship between molecular diffusion, turbulent diffusion and their mutual interference in non-reacting axisymmetric coaxial jets of variable Reynolds number. The reported investigation has been focused on the analysis of molecular diffusion of a He/CO_2 mixture in air by combining line imaging of Rayleigh scattering and laser Doppler anemometry (LDA) to determine length scales associated with differential diffusion and turbulent transport. Line imaging Rayleigh scattering was performed applying the index-matching method with a mixture of two gaseous species having scattering cross-sections respectively lower and higher of that of air and the cross-section of the mixture identical to that of the co-flowing air. Any measured variation in scattering intensity is therefore due to differential diffusion between the two species. Instantaneous and ensemble averaged line profiles of Rayleigh scattering intensity are presented and a characteristic length scale associated with differential diffusion is deduced. Autocorrelation analysis is applied to obtain the characteristic scale of differential diffusion fluctuations and the integral length scales of velocity fluctuations, as measured by LDA. Theoretical information from the literature is used in relating these scales to the molecular and turbulent diffusion coefficients, assuming homogeneous and isotropic turbulence, and the ratio of molecular to turbulent diffusivity is estimated as a function of the Reynolds number. The results confirm that the average contribution of molecular diffusion to the effective diffusivity into the air stream progressively reduces when the turbulence level increases. They also suggest that, at higher Re, the differential diffusion remains significant down to the scalar dissipation length scale, and could influence mixing at the molecular level and thus chemical reactions.

1. Introduction

The recognition that equal diffusivity of species is not a correct assumption in modelling turbulent diffusion flames was prompted by the early studies on jet flames of H_2 in air [1–4]. It was predicted that H_2 would mix faster than other species, due to its higher molecular diffusivity, at least at moderate Reynolds numbers. Several experimental and modelling papers deal with differential diffusion and its effects in combustion [5,6], and indicate that differences in species molecular diffusivities can have a significant impact on chemical evolution in turbulent non-premixed reacting flows. A recent paper [7] summarised, also from an historical perspective, the efforts made to isolate the effect of the differential diffusion from that of chemical reactions and heat release. It was suggested [4] to use non-reacting flows and Rayleigh scattering to visualise directly the presence of differential diffusion, by applying an index-matching imaging method with a mixture of two gaseous species such that its scattering cross-sections is identical to that of the surrounding air. Although only Raman scattering can add information on local absolute concentrations, the Rayleigh scattering has higher sensitivity and provides instantaneous images of the species relative distribution. Both one- and two-dimensional imaging measurements provided quantitative indication of differential diffusion in non-reacting jets [7–11] and its dependence on Reynolds number, although there is no consensus on a specific Re dependence.

Some experiments [5,8] have also shown that at higher Reynolds numbers, even when the ensemble averaged scattered signal does not show differential diffusion, the instantaneous species concentrations are affected by differential diffusion and suggest that this phenomenon remains important at the smallest scales of turbulent flows [7,8]. The persistence of the differential diffusion at higher Re supports the idea that differential diffusion can lead to significant effects on fluid composition and thus on chemical reactions, since they require mixing at the molecular level, where diffusive effects are significant [12]. This fact is a key point in the understanding of the combustion behaviour of hydrogen/hydrocarbons fuel

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Nomenclature

d	inner diameter of the fuel pipe, m
D_i	inner diameter of the air annulus, m
D_o	outer diameter of the air annulus, m
D_T^2	total effective displacement of infinitesimal fluid particle, m^2
L_E	integral length scale, m
M	momentum flux ratio of the inner to the outer jet
r	radial distance from the burner axis, m
R	normalized Rayleigh scattering intensity
R_L	Lagrangian correlation coefficient
Re	Reynolds number
Re_{L_E}	turbulent Reynolds number based on the integral length scale
Re_λ	Reynolds number based on the Taylor length scale
Sc	Schmidt number (ν/Γ)
T	time, s
T_E	Eulerian integral time scale, s
T_L	Lagrangian time scale, s
u	axial velocity fluctuation, m/s
$\bar{U}$	mean axial velocity component, m/s
U_i	inner jet bulk velocity, m/s
U_o	outer jet bulk velocity, m/s
w	turbulent velocity of a fluid particle, m/s
X	axial distance from the burner outlet, m
$\overline{Y^2}$	mean-square displacement due to turbulent diffusion, m^2

Greek letters

α	dimensionless constant in Eq. (10)
β	dimensionless constant in Eq. (3)
Γ	molecular diffusivity, m^2/s
Δ	interference term in Eq. (4), m^2
ε	coefficient of eddy diffusion, m^2/s
η	Kolmogorov length scale, m
η_B	Batchelor length scale, m
λ_D	scalar dissipation length scale, m
λ_T	Taylor length scale, m
Λ_L	scale of eddy diffusion, m
ν	kinematic viscosity, m^2/s
ξ	differential diffusion parameter
ρ_i	inner jet mixture density, kg/m^3
ρ_o	outer jet (air) density, kg/m^3
σ_i	scattering cross-section of species i, m^2
χ_i^0	initial mole fraction of species i

mixtures; indeed local non-uniformity in the fuel mixture composition, induced by the differential diffusion of hydrogen, could induce undesired effects on heat release, transport properties, extinction and pollutants formation.

Several papers have investigated the differential diffusion in non-reacting binary mixtures with hydrogen or helium injected in a low velocity co-flow of air and the jet Re has been varied from the laminar to the turbulent regime. The use of coaxial jets is widespread in the context of industrial burners for their ability to destabilize and mix the two fluid streams. The near-field of coaxial configurations is characterised by the momentum flux ratio of the inner to the outer jet $M = \rho_i U_i^2 / \rho_o U_o^2$ [13]. Coaxial jets at moderate momentum ratio offer the opportunity of simulating, at a fixed downstream location, the diffusive interpenetration of one stream into the other from the initial segregation up to the onset of molecular uniformity, by simply changing the value of M. This makes it possible to easily study how differential diffusion is affected by different regimes of turbulent diffusion.

Previous experiments involved only Rayleigh scattering measurements, precluding the possibility of determining the interaction of molecular diffusion with the local turbulent flow field. Without direct velocity measurements, the turbulent flow field has been only characterised by the jet Reynolds number. The aim of the present paper is to present an approach were laser Doppler anemometry (LDA) and Rayleigh scattering are used to estimate length scales associated with differential diffusion and turbulent transport in non-reacting axisymmetric coaxial jets of various Reynolds numbers. Molecular diffusion can easily be studied by means of scattering mechanisms sensitive to the molecules themselves and the Rayleigh scattering signal is a function of the local molecular density and the scattering cross-section. The tracer particles used by the LDA are best suited to characterise turbulent mixing, since they follow the fluid fluctuations and their displacement due to Brownian or molecular diffusion is completely negligible.

2. Experimental conditions and procedures

Line imaging of laser Rayleigh scattering was performed in a mixture of He and CO_2 in air, applying the index-matching imaging method [7,8]. The differential scattering cross-section of a gas mixture is given by the mole fraction-weighted sum of the cross-sections of the constituent species. Being the cross-sections of He and CO_2, relative to that of air at 532 nm and STP conditions, equal to 0.014 and 2.458 respectively [14], by mixing He and CO_2 in such proportion (mole fraction 59.5/40.5) that the mixture scattering cross-section is identical to that of air, any measured variation in the scattered intensity marks the presence of differential diffusion between the two components of the parent mixture.

Experiments were carried out using a non-premixed coaxial burner currently used in our laboratory to investigate combustion features of hydrogen/hydrocarbons fuel mixtures [15]. It consists of a vertical pipe (length 300 mm, inner diameter $d = 8$ mm) centred within a annular co-flow of air. The inner and the outer diameters of the air annulus are respectively $D_i = 15$ mm and $D_o = 36$ mm. The burner is confined in a closed chamber (200 mm inner diameter, 300 mm height), fitted with four opposite planar quartz windows, to reduce contamination of the ambient air and external flow disturbances. A schematic view of the experimental set-up is shown in Fig. 1. The experiments were conducted at ambient pressure and temperature, with the air flow accurately filtered to avoid Mie scattering by particulates that could overwhelm the Rayleigh scattering signal.

In the present experiments the inner jet was maintained at constant rate with $Re_{jet} = 4240$, based on the jet bulk velocity, pipe diameter and mixture kinematic viscosity $\nu = 2.15 \times 10^{-5}$ m^2/s. The air stream velocity was varied and, consequently, the Reynolds number, Re_{Air}, based on the annulus hydraulic diameter ($D_o - D_i$) and the bulk velocity of the air stream, varied between 2810 and 11380, as reported in Table 1. The corresponding momentum ratio, M, decreased from 21.03 to 1.28. The outer air stream accounted for about 81% of the total mass flux at the lowest flow rate and about 95% at the highest flow rate. The mixture of He and CO_2 was prepared in a mixing chamber and injected through the inner pipe ending flush with the outlet of the air stream. Accurate metering of all gases was performed using thermal mass flow controllers (accuracy ~1%). The constant He/CO_2 flow rate helped in maintaining perfect matching of the Rayleigh scattering cross-section of the jet mixture to that of air. Fine adjustment of the He/CO_2 ratio was