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ABSTRACT

The present study investigates the molecular unmixedness between the injected and entrained fluids in a turbulent jet as well as its dependence on the initial conditions. This unmixedness can be quantified as the parameter of molecular segregation between the ejecting "fuel" fluid (A) and the entrained "oxidizer" fluid (B), defined by $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$ (overbar denotes time-averaging). For the first time, an expression of the parameter has been derived for the two-fluid mixing in a heated turbulent nonreactive jet. That is, $\alpha = -\overline{\theta^2}/\left[\left(\Theta_o - \overline{\Theta}\right)\left(\overline{\Theta} - \Theta_a\right)\right]$, where Θ_o and Θ_a denote the ejected "warm" and entrained "cold" fluid temperatures, whereas Θ and θ are the instantaneous and fluctuating temperatures of the local fluid mixture. This expression of α is well validated by comparing the measured natural-gas flames from a smooth-contraction nozzle with those from a long-pipe nozzle. Moreover, the jet-nozzle configuration is found to show a strong effect on α. Likewise, the jet density ratio (R_ρ) is a highly influential factor: e.g., an increase in R_ρ reduces α substantially. In contrast, the effect of the jet-exit Reynolds number is less significant. In the present paper, we try to explain these observations.

I. INTRODUCTION

The turbulent mixing plays a crucial role in various practical applications such as combustors and chemical reactors. This physical process develops through three stages: i.e., large-scale entrainment, small to mid-scale dispersion, and molecular diffusion (Eckart[1]). The thorough mixing between two species A and B occurs only once their molecular diffusion has ensued. The segregation parameter $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$ (the overbar signifies the time-averaging), defined first by Danckwerts[2] for chemical reactions, is a critical parameter of measuring the unmixedness or incompleteness of mixing between A and B. Here, C and c represent the instantaneous and fluctuating concentrations and $C = \overline{C} + c$. Since $\overline{C_A C_B} = \overline{C}_A\overline{C}_B + \overline{c_A c_B} = \overline{C}_A\overline{C}_B(1+\alpha)$, the mean chemical reaction rate $R_{AB} \equiv \kappa\overline{C_A C_B}$ can be expressed as

$$R_{AB} = \kappa\overline{C}_A\overline{C}_B(1+\alpha), \tag{1}$$

where κ is the reaction-rate constant. If there is no molecular mixing at all between the species A and B, no reaction will occur so that $R_{AB} = 0$ and, thus, $\alpha = -1$. On the other hand, once the mixing has been fully completed, the resulting $\overline{C_A C_B} = \overline{C}_A\overline{C}_B$ and $\overline{c_A c_B} = 0$, hence the segregation parameter must be zero, i.e., $\alpha = 0$. It is deduced that a significant departure of α from zero measures the degree of incomplete mixing or the unmixedness. Correspondingly, the correlation $\overline{c_A c_B} \neq 0$ cannot be ignored. So, if the mean chemical reaction rate is calculated by Eq. (1) simply from the product $\overline{C}_A\overline{C}_B$ by taking $\alpha = 0$, serious errors will certainly occur in the computational fluid dynamics (CFD) modeling of turbulent combustion. Unfortunately, this is the way performed in many CFD calculations, as early criticized by Komori *et al.*[3]

Several previous experiments on the segregation parameter α were reported for turbulent flows involving the mixing and chemical reaction of two streams of initially unmixed reactants.[4–8] Those previous measurements[4–8] of $\overline{c_A c_B}$ and R_{AB} were made in moderately fast or slow reaction cases. Komori and Ueda[4] used a second-order chemical reaction between ozone (O_3) and nitric oxides (NOx), estimating α both in a reacting plume in grid-generated turbulence and in a reacting jet with a low-speed uniform coflow. Their estimation of α was not based on direct measurements of c_A and c_B but obtained from comparisons of the measured $\overline{C}_A$ and $\overline{C}_B$, with the numerical solutions of $\overline{C}_A$ and $\overline{C}_B$ in the mass-conservation equation. As a result, their approximate values of α were 20 and 1.5 in grid-generated turbulence and in a jet, respectively. To directly measure the concentration correlation $\overline{c_A c_B}$, Komori *et al.*[5] later conducted field measurements in a two-dimensional reacting plume of the atmospheric surface-layer flow (sea breeze). They arranged a line source with a length of 100 m at a vertical elevation of 1.5 m from the ground and emitted diluted NO from the source, with the concentration fluctuations of NO and O_3 to be simultaneously measured at a height of 1.5 m and downstream distances of 35 and 100 m from the line source. In this case, the measured α ranges from −0.025 to −0.27. Moreover, Mudford and Bilger[6] conducted experiments on reacting counter jets in a big smog chamber, directly measuring the concentration fluctuations of NO and O_3 by a gas-sampling technique. These investigators obtained that $\alpha = -0.01$ to −0.67. Later, Saetran *et al.*[7] measured α by the same technique at one streamwise location of a reacting mixing layer in grid turbulence and gained the segregation parameter of $\alpha \approx -0.25$ in the central region of the mixing layer. Interestingly, Bennani *et al.*[8] measured a liquid-phase chemical reaction between methylformate ($HCOOCH_3$) and sodium hydroxide (NaOH) in grid-generated turbulent flow with a high Schmidt number of $Sc = 700$, which is very different from the gaseous flows of $Sc \approx 1.0$.[5–7] They only measured the concentration of NaOH, indirectly estimating $\overline{c_A c_B}$ by assuming the mean velocity and concentration fields to be homogeneous over the cross section of a water tunnel. Their estimation is that $\alpha \approx -0.7$ throughout the measurement region. The magnitude of this α is rather greater than those obtained in the low-*Sc* flows of Mudford and Bilger[6] and Komori *et al.*[5]

The above values of α for the reactive turbulent flows vary greatly between −0.7 and 20. Such a significant variation is anticipated to result from experimental errors due to poor resolutions of the measurement probe, indirect methods of numerical simulations, and difficulties in performing these experiments under reacting conditions.[3] Indeed, Bilger *et al.*[9] explicitly disapproved the large positive values of α obtained by Komori and Ueda[4] and attributed them only to the measurement errors. They claimed, based on both their analysis and the measurements of Mudford and Bilger,[6] that α should be always negative in simple nonpremixed flows.

The claim of Bilger *et al.*[9] does not appear to coincide with the measurements of Tong and Warhaf[10] and Cai *et al.*[11] for the segregation parameter α between two species in nonpremixed and nonreacting turbulent jets. Tong and Warhaf[10] examined the turbulent mixing of two independently introduced thermal fields in a non-reacting turbulent jet. The two passive temperature sources were made by two heated fine-wire rings located differently and axisymmetrically in the flow, while an inference method (invoking the principle of superposition) was used to indirectly determine the correlation $\overline{c_A c_B}$ and, thus, $\alpha \equiv \overline{c_A c_B}/\overline{C}_A \overline{C}_B$. It was found that, ranging from −0.4 to 1.35, α initially depends strongly on the ring location and spacing; besides, the centerline α asymptotically approaches the value of 0.04 sufficiently downstream. Moreover, Cai *et al.*[11] investigated by experiment the scalar mixing in a turbulent jet, consisting of a center acetone-doped air jet (taking it as scalar 1) and an annular ethylene flow (scalar 2) together with an outer low-speed airflow (scalar 3). Using planar laser-induced fluorescence and Rayleigh scattering, they directly measured α whose centerline magnitude varies monotonically from −0.17 to the asymptotic value [≈0.045, see their Fig. 5(b)]. Obviously, the above α is not always negative but varies from −0.4 to some value >1.0. Both Tong and Warhaf[10] and Cai *et al.*[11] obtained a positive asymptotic-value along the jet centerline, inconsistent with the claim of Bilger *et al.*[9] for simple nonpremixed flows. Nevertheless, it is worth noting that the previous studies[10,11] concerned the mixing between two scalars ejected from two independent sources and advected by a turbulent jet flow. Such a mixing process differs from that between the original fluid issuing from a jet nozzle and that entrained from the surrounding environment. We investigate the latter case of mixing, whose segregation parameter will be approved fundamentally to be always negative in Sec. II.

The effectiveness of mixing of a turbulent jet with its surroundings is of great importance for a wide range of engineering and environmental applications such as industrial combustion (reactive) and pollutant dispersion (nonreactive) in the atmosphere. However, to the best of our knowledge, there have been no experimental data available for the segregation parameter or the unmixedness between the ejected and entrained fluids in any turbulent jet. If there were a relevant expression of α available for nonreacting jets, this gap in knowledge would be well filled by a large body of existing scalar data reported extensively in the literature, e.g., Refs. 10–24. This view has stimulated the present study that is to relate the parameter α to the existing scalar data for nonreactive turbulent jets.

Importantly, the impact of initial flow conditions on the scalar mixing field of a turbulent jet has been well investigated by the previous studies.[12–20] So, the same should be considered an important issue of the present research into the parameter α as well. The initial conditions of a turbulent jet are usually defined by the exit Reynolds number $Re \equiv U_o d/\nu$ (where U_o is the exit bulk velocity, d is the nozzle exit diameter, and ν is the kinematic viscosity), the exit radial profiles of mean velocity and turbulence intensity, and the global density ratio of the original-jet fluid (ρ_o) to ambient fluid (ρ_a), i.e., $R_\rho \equiv \rho_o/\rho_a$. Mi *et al.*[12] undertook an experimental investigation into the influence of initial velocity distributions on the passive scalar mixing field of a turbulent jet issuing from the round nozzle. They generated two sets of distinctly different initial velocity profiles using smooth-contraction (SC) and long-pipe (LP) nozzles. Their measurements of the passive scalar (temperature) field were conducted, using identical experimental facilities and a single measurement technique, in the slightly heated air jet from either SC or LP nozzle at $Re = 16\,000$ and $R_\rho \approx 1.0$. Mi *et al.*[12] found significant differences in the normalized profiles of the mean and RMS temperatures between the jets from the two nozzles throughout the near- to far-field region. They related the differences observed in the statistics of the scalar field to those in the two jets' underlying turbulence structure in the near field. A little bit later, Mi *et al.*[13] investigated the differences in mixing performance between axisymmetric turbulent jets issuing, respectively, from a SC nozzle, a LP and a sharp-edged orifice plate. They revealed that the jet issuing from the orifice plate provides the greatest rate of mixing with ambient fluid, while the LP jet has the lowest rate. Physical insight into the differences was explored using a planar imaging technique and measurements of power spectra of the fluctuating velocity.

On the other hand, Pitts[14,15] investigated the effects of *Re* and R_ρ on the centerline scalar mixing behavior of the turbulent jet issuing from a round LP nozzle. They claimed to find that the differences in *Re* and R_ρ do not influence the far-field statistical behavior of the jet. Richards and Pitts[16] later extended the investigation of Pitts[14,15] by varying both R_ρ and nozzle-type (SC and LP). These authors then concluded that the asymptotic state of the scalar field of a jet, as characterized by the mean spreading rate, the centerline mean decay rate, and the locally normalized RMS fluctuation (which they called the "unmixedness"), is not dependent on either the nozzle-type or R_ρ.

Apparently, there is some confliction between the findings of Mi *et al.*[12,13] and those of Pitts[14,15] and Richards and Pitts.[16] Namely, while the former[12,13] found a significant influence of the initial flow conditions on the jet's self-similar far-field scalar field, the latter[14–16] collectively claimed that the jet decays at the same rate, spreads at the same angle, and both the normalized mean and RMS scalars collapse in a form consistent with full self-similarity, regardless of the initial conditions. Naturally, a relevant question arises: whether or not do the initial conditions influence the segregation parameter in the self-preserving far-field region of a turbulent jet? In this context, the present study is specifically aimed at the following:

(1) Deriving the expression of α for the unmixedness between the ejected and entrained fluids in a nonreactive turbulent jet.
(2) Obtaining the α distributions of turbulent jets from the previous scalar measurements[10–18] based on the derived expression and then analyzing the obtained results.
(3) Clarifying the dependence of α on the initial conditions of the jet flow.

II. DERIVING THE EXPRESSION FOR $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$ IN A NONREACTING JET

Let C_A and C_B represent the instantaneous mass concentrations of the ejected original "warm" fluid of temperature Θ_o and the entrained ambient "cold" fluid of Θ_a in a turbulent nonreactive jet. Also, let Θ denote the mixture temperature in the jet. Assume the *same* fluid (e.g., air) to be used for both the ejected and entrained fluids, hence, with the *identical* specific heat. Under these conditions, the mass and energy balance equations can be expressed, respectively, as

$$C_A + C_B = 1 \tag{2}$$

and

$$C_A\Theta_o + C_B\Theta_a = \Theta. \tag{3}$$

A few manipulations of Eqs. (2) and (3) can obtain that

$$C_A = \frac{\Theta - \Theta_a}{\Theta_o - \Theta_a}, \quad C_B = \frac{\Theta_o - \Theta}{\Theta_o - \Theta_a}. \tag{4}$$

Then, their fluctuating components are

$$c_A = \frac{\theta}{\Theta_o - \Theta_a}, \quad c_B = -\frac{\theta}{\Theta_o - \Theta_a}, \tag{5}$$

where θ is the fluctuating temperature. It follows that the segregation parameter between the ejected and entrained fluids defined by $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$ can be attained from the temperature data, viz.:

$$\alpha = -\frac{\overline{\theta^2}}{(\Theta_o - \Theta)(\Theta - \Theta_a)}. \tag{6}$$

If we, like all the measured temperatures reported in the literature, use the relative mean temperatures above the ambient temperature (Θ_a), i.e., $\Theta_r = (\Theta - \Theta_a)$ and $\Theta_{ro} = (\Theta_o - \Theta_a)$, then after several manipulations, Eq. (6) can be rewritten as

$$\alpha = -\left(\frac{\theta'}{\Theta_r}\right)^2 \frac{\Theta_r}{\Theta_{ro} - \Theta_r}, \tag{7}$$

where $\theta' \equiv \overline{\theta^2}^{1/2}$ is the RMS temperature. For non-temperature scalars in jets, a similar relation to Eq. (7) for α can be easily obtained. For example, when the primary and ambient or coflow fluids are two different species (e.g., He/air, CH_4/air, etc.), the segregation parameter can be formulated as

$$\alpha = -\left(\frac{\overline{c_A^2}}{\overline{C}_A^2}\right)\frac{\overline{C}_A}{1 - \overline{C}_A}, \tag{8}$$

where C_A is the measured concentration or fraction of the species A issuing from a jet nozzle. Of note, Eqs. (2)–(8) should also apply if C_A and C_B represent the instantaneous mass concentrations of "warm" fluids from two thermal sources A and B in any turbulent flows (e.g., jets, wakes, …).

Here, it is worthwhile to make a few comments on the limiting and general cases of the segregation parameter α. While the thorough mixing between species A and B takes place, it must be that $\alpha = 0$ because C_A and C_B are always coexisting so that $\overline{C_A C_B} = \overline{C}_A\overline{C}_B$ and $\overline{c_A c_B} = 0$. On the other hand, the limiting case of no molecular mixing at all should correspond to $\alpha = -1$ for the following reason. If C_A and C_B do not coexist at any time and any point in space, the product of C_A and C_B must be zero, i.e., $C_A C_B = 0$. This yields that $\overline{C_A C_B} = \overline{C}_A\overline{C}_B + \overline{c_A c_B} = \overline{C}_A\overline{C}_B(1+\alpha) = 0$ and so that $\alpha = -1$, which is the limiting case of total segregation of species A and B. In general, the coexistence of A and B should take place occasionally or frequently, which means that $\overline{C_A C_B} > 0$ and $\overline{c_A c_B} < 0$. It follows that $(1+\alpha) > 0$ and $\alpha < 0$, or together $-1 < \alpha < 0$. Accordingly, the magnitude of $-\alpha$ can be regarded as the degree of segregation between A and B. Note that, if no chemical reactions occur, α should be always negative, as suggested by Eqs. (7) and (8). When chemical reactions take place, Eq. (2) must be invalid. So, Bilger *et al.*[9] deduced that α should be positive for some cases although being negative for most reactive cases. In addition, for the nonreacting multi-scalar mixing or the mixing of two species A and B advected by a single turbulent flow (e.g., jet), it is equally likely for α to be positive (though mainly negative) because the inherent relations $C_A + C_B = 1$ and $c_A + c_B = 0$ for the two-scalar mixing do not hold in the mixing of multiple (>2) scalars.

III. RESULTS AND DISCUSSION

A. The turbulent mixing of the original-ejected and entrained fluids in a slightly heated jet

The mean and RMS scalar distributions for turbulent nonreactive jets have been reported quite extensively, e.g., in Refs. 10–24. However, the available data are scattered from paper to paper due to distinct experimental (initial/boundary) conditions and different setup and measurement devices used for respective measurements of turbulent jet flows.[12] This study investigates the segregation or the incompleteness of turbulent mixing between the original-ejected and ambient-entrained fluids mainly in a circular jet. So, to properly and accurately calculate the segregation parameter α from Eq. (7), we choose the mean and RMS temperatures (Θ_r and θ') of a slightly heated circular jet of Mi *et al.*[12] and a slightly heated planar jet of Browne *et al.*[17] Of note, these two datasets have been highly cited and their reliability and comparability are sufficiently good; see the original papers[12,17] for details of their experimental settings and jet nozzle geometry conditions. In addition, the correctness of the presently obtained α in the circular jet is confirmed, and so Eq. (7) is validated, by combustion experiments of Langman *et al.*[18] who measured the natural-gas jet flames from circular nozzle burners. The obtained results are shown below.

Figure 1 compares the centerline variations of the segregation parameter and the normalized mean and RMS temperatures (i.e., α, Θ_{rc}/Θ_{ro}, and θ_c'/Θ_{rc} vs x/d) for a long-pipe (LP) jet with those for a smooth-contraction (SC) jet; here, d and x are, respectively, the downstream distance from and the diameter of the nozzle exit. On the plot, we also present the variable $\eta = (\alpha_{LP} - \alpha_{SC})/\alpha_{SC}$ to show more clearly the difference in α between the LP and SC jets. Both jets were measured at $Re = 16\,000$. Note also that the data of Θ_{rc}/Θ_{ro} and θ_c'/Θ_{rc} are reproduced from Mi *et al.*[12] It is clearly demonstrated that the mean temperature Θ_{rc} decays at a lower rate in the LP jet than in the SC jet, especially at $x/d < 30$; concurrently, the LP jet spreads more slowly (not shown here but reported in Ref. 12). This is because the underlying structures are more coherent or organized in the near and transition regions of the latter flow. Correspondingly, the RMS value along the centerline is higher in the SC jet.

Figure 1 also illustrates the centerline variations of α of the two jets, estimated from Eq. (7) and the data of Θ_{rc}/Θ_{ro} and θ'/Θ_{rc}. It is observed that the centerline α is always closer to zero in the LP jet than the SC jet. This difference is demonstrated more obviously by the centerline η curve. Interestingly, η grows from nearly zero at the exit ($x = 0$) to about 1.0 around the end of the SC-jet's potential core and then drops to 0.3–0.4 in the far field. Such a centerline variation of η is not difficult to be understood when considering the "laminar" and "turbulent" states of the initial SC and LP jets. However, it is still surprising that the LP-jet unmixedness ($-\alpha_{LP}$) is smaller than the SC-jet one ($-\alpha_{SC}$) by 30%–40% in the far field. This suggests that the molecular mixing in the SC jet is considerably poorer than the LP jet even in the far field.

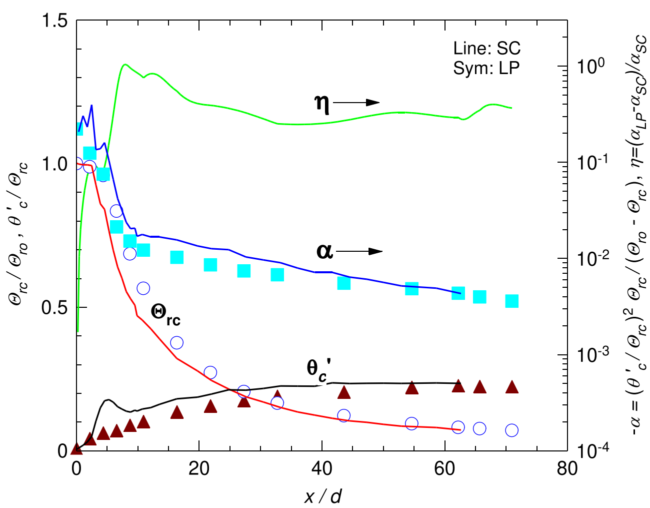

FIG. 1. Variations of the segregation parameter α (——, the blue square) and its difference η between the LP and SC jets (——), the normalized mean temperature Θ_{rc}/Θ_{ro} (——, the blue circle), and the normalized RMS fluctuation θ_c'/Θ_{rc} (——, the brown triangle) in the SC and LP jets both for $Re = 16\,000$.[12] Note: the logarithm is taken on α and η.

Figure 2 shows the radial profiles of α, η, Θ_{rc}/Θ_{ro}, and θ'/Θ_{rc} vs $r/(x\text{-}x_o)$, where x_o is the x-location of the virtue origin, obtained in the self-preserving far-field LP and SC jets. What we can learn from this plot is clear: i.e., like the centerline case, the magnitudes of $-\alpha$ and the normalized RMS fluctuation θ'/Θ_{rc} across the far-field LP jet are generally smaller than those of the SC counterpart. In particular, the radial profile of $\eta = (\alpha_{LP} - \alpha_{SC})/\alpha_{SC}$ indicates that the unmixedness is substantially greater across the SC jet than the LP jet. On average, the value of $-\alpha$ for the SC jet is about 36% higher than that of the LP jet; note that the averaging is taken over the range of $0 < r/(x\text{-}x_o) < 0.16$.

Based on the above differences in α and θ'/Θ_{rc} between the two jets, it is hypothetically suggested that the LP jet has less unmixedness or more thorough mixing at a molecular level between the original-ejected and ambient-entrained fluids. However, this appears to be the opposite of the claim of Mi *et al.*[12,13] that the LP jet should be globally mixed by the ambient flow at a lower rate because the mean scalar field of the SC jet was found to both decay and spread more rapidly. Such a contradiction can be clarified as follows. According to Eckart,[1] the turbulent mixing is a three-stage process: the first is large-scale entrainment, the second is smaller-scale dispersion, and the final is molecular diffusion. Careful inspections of Figs. 1 and 2 suggest that the SC jet is more effective in the first stage (i.e., the claim of Mi *et al.*[13]) due to the entrainment enhanced by more highly coherent structures, while the LP jet performs better in the final stage (i.e., molecular diffusion) of the turbulent mixing process. Note that both $-\alpha$ and θ'/Θ_{rc} are smaller in the LP jet than in the SC jet. This seems to support the conventional view that the magnitude of θ'/Θ_{rc} represents the unmixedness of the turbulent jet, as previously often claimed (e.g., Refs. 14–16). Nevertheless, this may not be the case when comparing the results of a circular SC jet with those of a planar SC jet (Fig. 5).

In fact, the better molecular mixing of the LP jet than the SC jet has been demonstrated by Langman *et al.*[18] using natural-gas jet

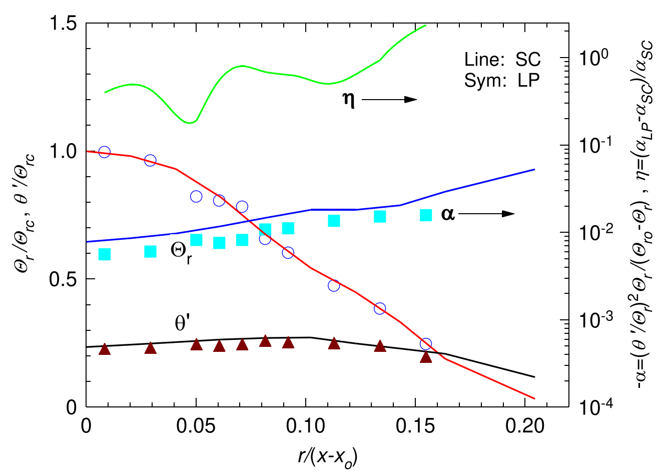

FIG. 2. Radial profiles of the segregation parameter α (——, the blue square) and its relative difference η between the LP and SC jets (——), the relative mean temperature Θ_r/Θ_{rc} (——, the blue circle), and the relative RMS fluctuation θ'/Θ_{rc} (——, the brown triangle) in the far-field SC and LP jets both for $Re = 16\,000$.[12] Note: the logarithm is taken on α and η.

flames issuing from the LP and SC nozzles of $d = 5$ mm. Figure 3 compares their measurements for the normalized flame length (L_F/d) and radiant fraction (χ_r) of the LP flame against U_o (ejecting bulk-mean velocity) with those of the SC flame. Here, L_F is the mean length obtained by averaging the instantaneous lengths of time-recording flame images, whereas $\chi_r = Q_r/Q_F$ with Q_r being the total flame radiated power (kW) and Q_F the input power (kW). Note also that all the data are reproduced from Figs. 3 and 7 of Langman *et al.*[18] It is evident that, as U_o rises, L_F gradually increases and χ_r decreases for both SC and LP flames at $U_o \leq 46$ m/s. When $U_o > 46$ m/s, both L_F and χ_r vary little for each flame, but the magnitudes of L_F for the two flames differ discernibly. In particular, at $U_o \leq 46$ m/s, the LP flame's length and radiation fraction are both smaller than those of the SC flame. More specifically, the average values of L_F/d and χ_r are approximately 148.6 and 11.3% for the LP flame vs 156.5 and 12.2% for the SC flame. These differences approve the better molecular mixing between the original-ejected and ambient-entrained fluids in the LP jet than in the SC jet, consistent with the implication gained from the difference in α between the two jets. The explanation follows. When the molecular mixing between the ejected fuel and ambient oxidant becomes poorer, the complete combustion will need a longer time and, thus, a larger flame volume, consequently with a higher radiation due to more generation of soot. Of note, soot dominates the radiant heat transfer in gaseous flames. In other words, relative to the LP flame, the larger volume and higher radiation of the SC flame result certainly from its poorer molecular mixing with the ambient flow. Therefore, the value of $-\alpha$ should be greater for the SC jet than for the LP jet. Indeed, this is the case shown above in Figs. 1 and 2.

Now, a new look is given into the radial variation of the segregation parameter α. Figure 4 illustrates the radial profiles of α obtained in the LP jet ($x/d = 15$–65) and SC jet ($x/d = 8$–60) both for $Re = 16\,000$.[12] It is observed that the segregation parameter reduces either as the flow proceeds downstream or as the radial distance (r) from the centerline decreases. These observations are expected because the local mixedness in each jet must grow with increasing x and decreasing r. However, unlike those of Θ_r/Θ_{rc} and θ'/Θ_{rc}, the radial profiles of α at different x-locations in the far field do not collapse onto a single curve or become self-similar, which might not be expected from the first thought. Likewise, the centerline variation of α does not seem to approach self-preserving as observed from Fig. 1. Here, we explain these results below. Equation (7) can be re-expressed as

$$\alpha = -\left(\frac{\theta'}{\Theta_{rc}}\right)^2 \left(\frac{\Theta_{rc}}{\Theta_r}\right)\left(\frac{\Theta_{ro}}{\Theta_{rc}} - \frac{\Theta_r}{\Theta_{rc}}\right)^{-1}. \quad (9)$$

In the far-field self-similar region of a circular turbulent jet, the self-similar relations of $\Theta_{ro}/\Theta_{rc} = a(x/d) + b$, $f(\eta) = \Theta_r/\Theta_{rc}$, and $g(\eta) = \theta'/\Theta_{rc}$ should be valid. Their substitutions into Eq. (9) lead to

$$\alpha(\eta) = -g^2(\eta) f^{-1}(\eta)\left[K\left(\frac{x - x_o}{d}\right) - f(\eta)\right]^{-1}, \quad (10)$$

where $f(\eta)$ and $g(\eta)$ are the self-similar functions with $\eta = r/(x\text{-}x_o)$ and x_o being the x-location of the virtual origin; K is a constant. Equation (10) indicates that the radial profiles of α cannot collapse onto a single curve in the far field but always decreases with increasing x, fully consistent with Fig. 4. However, on the centerline, Eq. (10) can be simplified as

$$\alpha(0) = -g^2(0)\left[K(x - x_o)/d - 1\right]^{-1}, \quad (11)$$

where $g(0)$ is invariable with x. Such a simplified relation has gained a strong support from Fig. 5 where, as indicated on the plot, a line of $\log(-\alpha)$ vs x/d occurs well at $x/d > 10$ for the circular SC jet.

In the above, we have investigated the SC and LP jets that initially have identically circular shapes in cross section but differ in their nozzle configurations, i.e., a smoothly contracting (SC) cross section vs a constant cross section long-pipe (LP). Next, a similar investigation is conducted on initially differently shaped jets. Figure 5 compares on-centerline variations of α, Θ_{rc}/Θ_{ro} and θ_c'/Θ_{rc} for the circular SC jet ($Re = 16\,000$) with those for a planar SC jet ($Re = 7620$) studied by Browne *et al.*[17] Note that the planar jet is injected from a long (rectangular) slot exit nozzle which differs substantially from the circular one. It is evident that the mean temperature decays at a significantly higher rate in the circular jet than in the planar jet. This suggests that the former jet entrains and mixes with ambient fluid at greater rates. Figure 5 also demonstrates that θ_c'/Θ_{rc} approaches asymptotically to a far-field value being considerably higher in the circular than in the planar jet. Moreover, α is always closer to zero for the circular jet than for the planar counterpart. This contrasts strikingly to a higher asymptotic value of $g(\eta = 0)$ or θ_c'/Θ_{rc} for the circular jet against the planar counterpart (see Fig. 5). Hence, the circular jet should be more effective not only in achieving the thorough mixing molecularly but also in large-scale entrainment. It is suggested, too, that a smaller value of the far-field θ_c'/Θ_{rc} in the planar jet is not related to a smaller unmixedness of this jet against the circular jet. Moreover, unlike that of α, the magnitude of θ_c'/Θ_{rc} does not appropriately measure the unmixedness, which was often claimed previously, e.g., by Pitts.[14,15]

In addition, there is a self-evident finding to be made from Figs. 1 and 5. Namely, an unconfined jet flow can never achieve $\alpha = 0$ so that the truly thorough mixing is never achieved between the nozzle ejecting fluid and the entrained ambient fluid. This results inevitably from the free jet, an open flow system, into which some "fresh" fluid is continuously entrained. In fact, we can also explain this through Eq. (10).

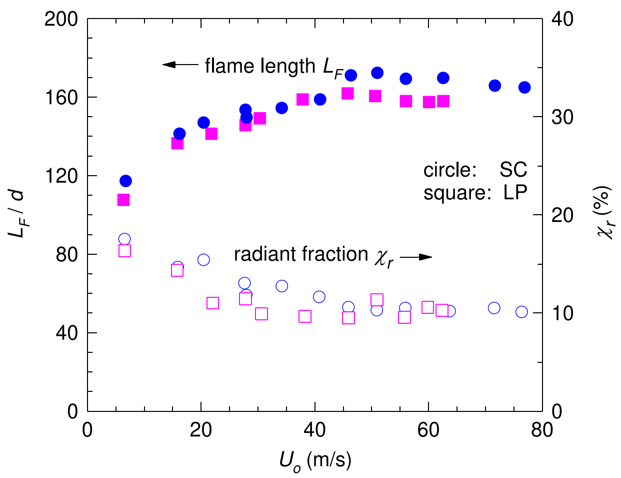

FIG. 3. Flame lengths (L_F) and radiant fractions (χ_r), vs bulk-mean exit velocity U_o, of two natural gas jet flames issuing from the 5 mm LP and SC nozzles.[18]

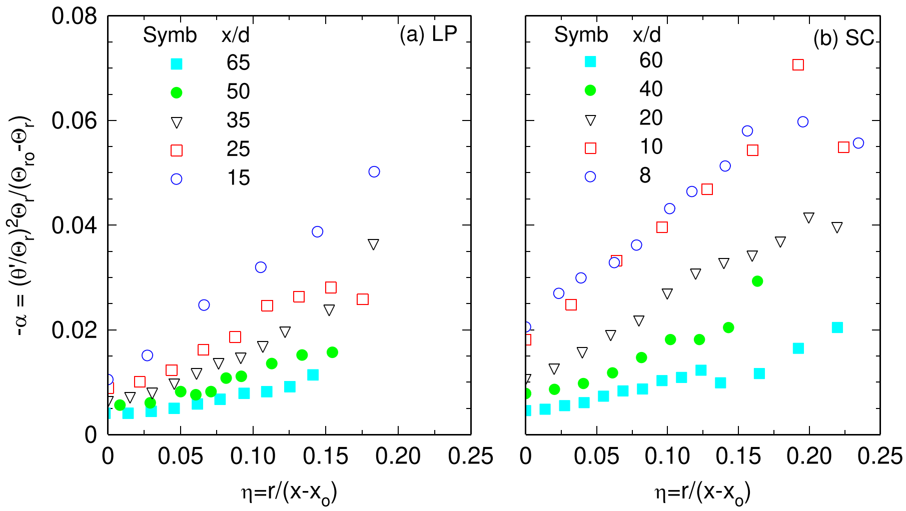

FIG. 4. Radial profiles of the segregation parameter α at different indicated values of *x/d* in (a) the LP jet and (b) the SC jet both for $Re = 16\,000$.[12]

When the jet develops downstream into the self-similar region in the far field, θ_c'/Θ_{rc} (and, thus, $\overline{\theta_c^2}/\Theta_{rc}^2$) will become approximately constant, see Fig. 5. However, the case of $\alpha = 0$ occurs only when $x \to \infty$, which is practically impossible.

B. Three-scalar mixing in turbulent round jets vs jet-ambient mixing

As noted in Introduction, Bilger *et al.*[9] claimed that that the segregation parameter (α) must be negative in nonpremixed turbulent flows, regardless of being reactive or nonreactive. This point has been analytically approved by the present work (Sec. II) for the two-stream case of an ejected stream and an ambient flow (together forming a jet). However, this does not seem to apply for the cases of Tong and Warhaf[10] and Cai *et al.*[11] for a nonpremixed and nonreacting turbulent jet when judging based only on their α measurements. Tong and Warhaf[10] examined the turbulent mixing of two independently introduced thermal fields in a circular SC jet. The two passive temperature sources A and B were made by two heated fine-wire rings located differently and axisymmetrically in the flow. These authors used a chromal-constant thermocouple to measure the mean temperature and platinum-resistant wires of diameter 1.25 μm to measure the fluctuating temperature. Of note, they employed an indirect inference method to obtain $\overline{c_A c_B}$ and, thus, $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$. They obtained that $\alpha = -0.4$ to 1.35 and also that the centerline $\alpha \to 0.04$ at $x/d > 15$. Actually, the turbulent mixing of Tong and Warhaf[10] does not belong to the two-scalar mixing of the ejected original and entrained ambient fluids. Instead, it was a three-scalar mixing case: i.e., two temperature scalars from the two heated rings A and B plus a lower temperature from the jet fluid mixture.

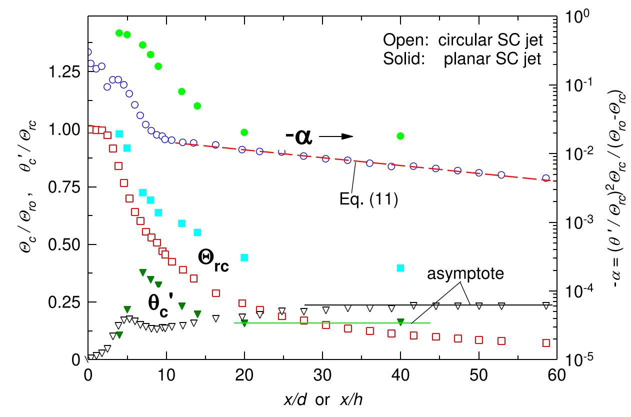

FIG. 5. Variations of the segregation parameter α (the blue and green circle), the normalized mean temperature Θ_{rc}/Θ_{ro} (the red and blue square) and the normalized RMS fluctuation θ_c'/Θ_{rc} (the open inverted triangle and the green triangle) in a turbulent circular jet for $Re = 16\,000$.[12] (the blue circle, red square, and open inverted triangle) and a planar jet for $Re = 7620$[17] (the green circle, blue square, and green inverted triangle). Note: the logarithm is taken on α.

Sixteen years later, Tong's group, i.e., Cai *et al.*,[11] conducted another investigation on the three-scalar mixing of turbulent jets. Namely, they studied the turbulent mixing between a center acetone-doped air jet (regarded as scalar 1 with C_1 representing the concentration in the mixture, the initial velocity of 34.5 m/s), an annular ethylene flow (scalar 2 with C_2, 32.5 m/s), and an outer low-speed airflow (scalar 3 with C_3, 0.4 m/s). They used the techniques of planar laser-induced fluorescence and Rayleigh scattering to measure the instantaneous concentrations C_1 and C_2, hence directly obtaining the segregation parameter $\alpha_0 \equiv \overline{c_1 c_2}/\overline{C}_1\overline{C}_2$. It was found that the centerline α_0 increases monotonically from -0.17 to about 0.045 (perhaps the asymptotic value), as shown in Fig. 5(b) or Fig. 6. Below, we explain why some positive values of the parameter could be gained by Tong and Warhaf[10] and Cai *et al.*[11] for their mixing cases in a nonreacting turbulent jet.

For the three-scalar mixing, the following relations must be valid:

$$\text{(Instantaneous)}\ C_1 + C_2 + C_3 = 1, \tag{12a}$$

$$\text{(Averaging)}\ \overline{C}_1 + \overline{C}_2 + \overline{C}_3 = 1, \tag{12b}$$

$$\text{(Fluctuating)}\ c_1 + c_2 + c_3 = 0. \tag{12c}$$

A couple of manipulations on Eq. (12c) can lead to $2c_1c_2 = c_3^2 - (c_1^2 + c_2^2)$ and, thus,

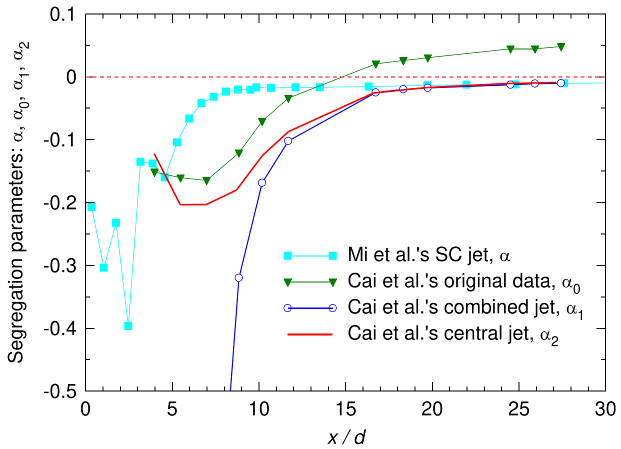

FIG. 6. Centerline variations of the segregation parameters α_0 directly measured by Cai *et al.*,[11] α_1 and α_2 estimated from Eq. (8) and Cai *et al.*,[11] and α from Eq. (7) and Mi *et al.*[12]

$$\overline{c_1c_2} = \left[\overline{c_3^2} - \left(\overline{c_1^2}+\overline{c_2^2}\right)\right]\Big/2. \tag{13}$$

Equation (13) implies that $\overline{c_1c_2}$ will become positive as x is sufficiently large for the mixing cases of Tong and Warhaf[10] and Cai *et al.*[11] The reason follows. As x increases, both C_1 and C_2 reduce while C_3 rises and so do the related mean concentrations. Consequently, $(\overline{c_1^2}+\overline{c_2^2})$ decreases and becomes smaller than $\overline{c_3^2}$ sufficiently downstream, hence leading to $\alpha_0 \equiv \overline{c_1c_2}/\overline{C}_1\overline{C}_2 > 0$. This, as demonstrated in Fig. 6, approves the appropriateness of the positive results of Tong and Warhaf[10] and Cai *et al.*[11]

In fact, it is also reasonable to treat the downstream case of Cai *et al.*[11] as the two-scalar mixing between the combined jet (a central acetone-doped air jet + an annular ethylene flow) and the outer low-speed airflow. In this case, based on the definition and Eqs. (12a)–(12c), the segregation parameter can be expressed as

$$\alpha_1 = \frac{\overline{c_3(c_1+c_2)}}{\left[\overline{C}_3\left(\overline{C}_1+\overline{C}_2\right)\right]} = -\frac{\overline{(c_1+c_2)^2}}{\left[1-\left(\overline{C}_1+\overline{C}_2\right)\right]\left(\overline{C}_1+\overline{C}_2\right)}, \tag{14}$$

which is always negative. Note that Eq. (14) is identical with Eq. (8). Based on Eq. (14), α_1 can be estimated using the data of Cai *et al.*[11] shown in their Figs. 4 and 5. Figure 6 displays the centerline results of α_0 and α_1 vs the data of α shown in Figs. 1 and 5 for the slightly heated SC jet of Mi *et al.*[12] In addition, we may obtain $\alpha_2 = -\overline{c_1}^2/\left[\overline{C}_1\left(1-\overline{C}_1\right)\right]$ [i.e., Eq. (8)] by considering the downstream case of Cai *et al.*[11] as the two-scalar mixing between the central acetone-doped air jet and the outer flows (i.e., the combined annular ethylene flow and outer airflow). The result is also shown in Fig. 6. Evidently, in the jet of Cai *et al.*, α_1 and α_2 become nearly identical at $x/d \geq 16$, whereas $\alpha_1 < \alpha_2$ at $x/d < 16$. These two observations can be easily understood. The first is due to the sufficient distance of mixing of $x \geq 16d$ over which the outer airflow has well reached the jet centerline so that both negative α_1 and $\alpha_2 \to 0$. The second observation is valid because the entrained annular ethylene reached the centerline much earlier than the coflowing air.

There is another significant observation from Fig. 6: namely, the negative centerline segregation parameter approaches zero far more rapidly in the circular jet of Mi *et al.*[12] than in that of Cai *et al.*[11] In other words, the efficiency of mixing of the former jet with its surrounding was substantially higher than that of the latter jet. This can be well explained, too, when considering the difference in boundary conditions between the two jets. While the SC jet of Mi *et al.*[12] issued into a still surrounding (to be called perhaps "zero-speed coflow"), the one of Cai *et al.*[11] was accompanied with a low-speed (0.4 m/s) coflow of air. Numerous previous studies, e.g., Refs. 25–27, have demonstrated a very strong effect of the coflow speed on the jet mixing and flame stability. For example, Dahm and Dibble[25] found that an increase of just 1% coflow velocity could result in a 50% reduction in the flame blow-out velocity. Likewise, Han and Mungal[27] revealed that slowing down the coflow could increase entrainment in a manner exponentially dependent on the density-weighted velocity ratio of jet to coflow.

C. Effects of jet's initial conditions on the turbulent unmixedness

According to the previous work (e.g., Refs. 12, 13, 18, and 28–30), the flow initial conditions should play a significant role in affecting the turbulent unmixedness of any flows all the way from near to far field. Indeed, Figs. 1–5 together demonstrate that a geometric variation of jet nozzle significantly influences the downstream parameter α. Specifically, as a long pipe (LP) is changed to a smooth-contraction (SC) nozzle, the SC jet is more effective in large-scale turbulent mixing due to more highly coherent structures, while the LP jet performs better in molecular diffusion of the turbulent mixing process. Differently, when the nozzle shape is varied from a circle to a very long slot rectangle, the circular jet appears to accomplish both molecular diffusion and large-scale entrainment more effectively than the planar jet.

Next, we investigate the dependence of the centerline α on both the Reynolds number Re and the density ratio R_ρ. The use of Eq. (8) enables the calculations of α from the concentration measurements of Pitts[14,15] and Dowling and Dimotakis.[19] Figure 7 illustrates the centerline variations of α for different Re jets of C_3H_8/air by Pitts[14] with $R_\rho = 1.55$ and those of C_2H_4/N_2 and C_3H_6/argon by Dowling and Dimotakis[19] with $R_\rho \approx 1.0$. To see the Re effect more clearly, three best-fit curves are drawn on the plot. Careful comparisons suggest that the segregation parameter rises as Re increases. This Re effect appears to be significant for low Re but weakens rapidly as Re grows. Specifically, the effect of Re on α, obtained from Pitts,[15] is discernible from $Re = 3960$ to $Re = 7930$ but becomes negligible at higher Re. Based on the data of Dowling and Dimotakis,[19] the Reynolds number effect is obviously negligible at $Re > 5000$.

Figure 7 also shows a clear distinction between the α values from Dowling and Dimotakis[19] and those from Pitts.[15] This distinction does not result from different values of Re but from those of R_ρ. To examine the effect of R_ρ, Fig. 8 shows the centerline values of α for different gas-pair jets whose R_ρ grows greatly from 0.14 to 5.11. Here, α is estimated via Eq. (8) from Figs. 2 and 4 of Pitts.[14] Figure 8 clearly demonstrates that the segregation parameter reduces substantially as the density ratio rises. This reduction weakens with increasing the downstream distance. However, the difference in α between different density-ratio jets does not appear to vanish even in the very far field.

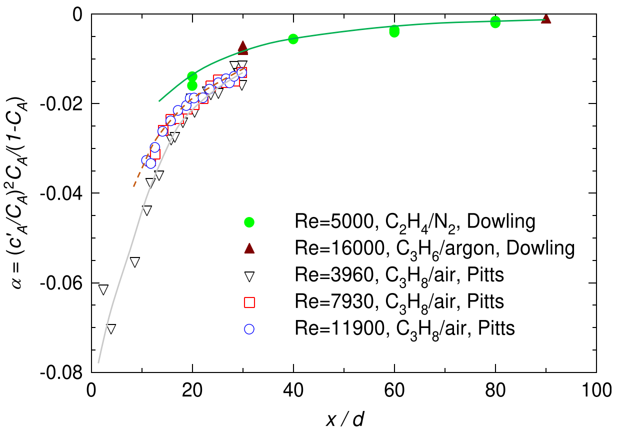

FIG. 7. Variations of the segregation parameters of different Reynolds number (*Re*) jets, estimated from Dowling and Dimotakis[19] with $R_\rho \approx 1.0$ and Pitts[15] with $R_\rho = 1.55$. Symbols are for the values obtained from the relevant concentrations,[15,19] while curves are the best-fit results.

It is certainly implied that the low-density jet is more efficiently mixed with the ambient fluid than the high-density jet.

IV. CONCLUDING REMARKS

(1) In the present work, we have for the first time derived the expression (7) for the parameter $\alpha \equiv \overline{c_A c_B}/\overline{C}_A\overline{C}_B$ of molecular segregation between the original-ejected "warm" fluid (A) and the entrained ambient "cold" fluid (B) in a turbulent nonreacting jet, which belongs to the two-scalar mixing. The limiting case of $\alpha = 0$ represents the thorough mixing between A and B while that of $\alpha = -1$ corresponds to no molecular mixing at all. For the two-scalar mixing, frequently A and B coexist so that $C_A C_B > 0$ and $c_A + c_B = 0$. It follows that both $\overline{C_A C_B} = \overline{C}_A\overline{C}_B(1+\alpha) > 0$ and $\overline{c_A c_B} = \alpha\overline{C_A C_B} < 0$ are valid; hence, $-1 < \alpha < 0$. However, for the multiple (>2) scalar mixing, the parameter α cannot always (but mainly) remain negative because $c_A + c_B \neq 0$. Importantly, the magnitude of α reflects the extent of molecular unmixedness between A and B.

(2) The parameter α is related by Eq. (7) or (8) to the scalar mean ($\overline{C}$) and RMS ($c' = \overline{c^2}^{1/2}$). This parameter has been found to be more appropriate than $c'/\overline{C}$, i.e., the conventional "unmixedness," to represent the real unmixedness between the original-ejected and entrained-ambient fluids.

(3) The correctness of the expression (7) or (8) has been validated experimentally by comparing the natural-gas flame from a smooth-contraction (SC) nozzle with that from a long-pipe (LP) nozzle. More specifically, relative to the LP case, the SC jet exhibits a greater value of $-\alpha$, i.e., less thoroughly mixing with ambient, thus resulting in a larger flame with a higher radiation fraction from the SC nozzle burner.

(4) The dependence of the parameter α on the initial jet conditions has been demonstrated to be generally significant all the way from near to far field of a jet flow. Although the Reynolds number effect on α is not very significant, the density ratio of jet-to-ambient fluids plays an important role in affecting α. Besides, the geometric variation of nozzle configuration strongly influences the magnitude of α over the entire jet flow field.

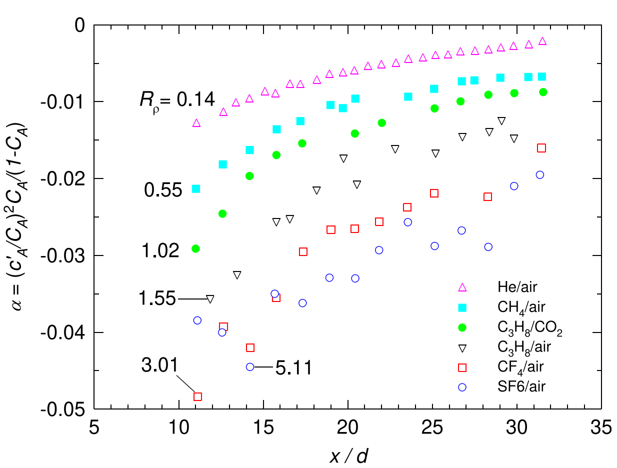

FIG. 8. Variations of the segregation parameters of different (density ratio R_ρ) gas-pair jets at $Re \approx 7900$ for SF6/air and $Re \approx 4000$ for others, estimated from Eq. (8) and Pitts.[14]

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Jianchun Mi: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal); Writing – review & editing (equal). **Mengwei Wu:** Data curation (equal); Formal analysis (equal); Investigation (equal). **Minyi Xu:** Data curation (equal); Formal analysis (equal); Investigation (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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