On molecular unmixedness between the ejected original and entrained ambient fluids in a turbulent jet

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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\textsuperscript{1}). The thorough mixing between two species A and B occurs only once their molecular diffusion has ensued. The segregation parameter \( \varepsilon \equiv \overline{c_A c_B} / \overline{C_A C_B} \) (the overbar signifies the time-averaging), defined first by Danckwerts\textsuperscript{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 \( C = C + c \). Since \( \overline{c_A c_B} = \overline{C_A C_B} + \varepsilon \overline{c_A C_B} = \overline{C_A C_B}(1 + \varepsilon) \), the mean chemical reaction rate \( R_{AB} \equiv \kappa \overline{c_A C_B} \) can be expressed as

\[
R_{AB} = \kappa \overline{c_A C_B}(1 + \varepsilon), \tag{1}
\]

where \( \kappa \) 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, \( \varepsilon = -1 \). On the other hand, once the mixing has been fully completed, the resulting \( \overline{c_A c_B} = \overline{C_A C_B} \) and \( \varepsilon \overline{c_A c_B} = 0 \), hence the segregation parameter must be zero, i.e., \( \varepsilon = 0 \). It is deduced that a significant departure of \( \varepsilon \) from zero measures the degree of incomplete mixing or the unmixedness. Correspondingly, the correlation \( \varepsilon \overline{c_A c_B} \neq 0 \) cannot be ignored. So, if the mean chemical reaction rate is calculated by \( \overline{R}_{AB} \) simply from the product \( \overline{C_A C_B} \) by taking \( \varepsilon = 0 \), 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.\textsuperscript{3}

Several previous experiments on the segregation parameter \( \varepsilon \) were reported for turbulent flows involving the mixing and chemical reaction of two streams of initially unmixed reactants.\textsuperscript{4-8} Those previous measurements of \( \varepsilon \overline{C_A C_B} \) and \( R_{AB} \) were made in moderately fast or slow reaction cases. Komori and Ueda\textsuperscript{7} used a second-order chemical reaction between ozone (O\textsubscript{3}) and nitric oxides (NO\textsubscript{x}), estimating \( \varepsilon \) both in a reacting plume in grid-generated turbulence and in a reacting jet with a low-speed uniform coflow. Their estimation of \( \varepsilon \) 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 \( \varepsilon \) were 20 and 1.5 in grid-generated turbulence.
and in a jet, respectively. To directly measure the concentration correlation $c_{17/C_0}$, 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 $x$ ranges from $-0.025$ to $-0.27$. Moreover, Mudford and Bilger\(^11\) 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 $x = -0.01$ to $-0.67$. Later, Saetran et al.\(^7\) measured $x$ by the same technique at one streamwise location of a reacting mixing layer in grid turbulence and gained the segregation parameter of $x \approx -0.25$ in the central region of the mixing layer. Interestingly, Bennani et al.\(^3\) measured a liquid-phase chemical reaction between methylformate (HCOOCH$_3$)/C$_17$ 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 $Sc \approx 1.0$.\(^5\) They only measured the concentration of NaOH, indirectly estimating $c_{17/C_0}$ by assuming the mean velocity and concentration fields to be homogeneous over the cross section of a water tunnel. Their estimation is that $x \approx -0.7$ throughout the measurement region. The magnitude of this $x$ is rather greater than those obtained in the low-$Sc$ flows of Mudford and Bilger\(^2\) and Komori et al.\(^3\).

The above values of $x$ 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.\(^11\) explicitly disapproved the large positive values of $x$ obtained by Komori and Ueda\(^1\) and attributed them only to the measurement errors. They claimed, based on both their analysis and the measurements of Mudford and Bilger,\(^2\) that $x$ should be always negative in simple nonpremixed flows.

The claim of Bilger et al.\(^1\) does not appear to coincide with the measurements of Tong and Warhaf\(^9\) and Cai et al.\(^11\) for the segregation parameter $x$ between two species in nonpremixed and nonreacting turbulent jets. Tong and Warhaf\(^9\) 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 $c_{17/C_0}$ and, thus, $x \equiv c_{17/C_0}/C_17C_0$. It was found that, ranging from $-0.4$ to $1.35$, $x$ initially depends strongly on the ring location and spacing; besides, the centerline $x$ 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 $x$ whose centerline magnitude varies monotonically from $-0.17$ to the asymptotic value [$x=0.045$, see their Fig. 5(b)]. Obviously, the above $x$ is not always negative but varies from $-0.4$ to some value $>1.0$. Both Tong and Warhaf\(^9\) and Cai et al.\(^11\) obtained a positive asymptotic-value along the jet centerline, inconsistent with the claim of Bilger et al.\(^1\) for simple nonpremixed flows. Nevertheless, it is worth noting that the previous studies\(^11\) 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 $x$ 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 $x$ 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 $x$ as well. The initial conditions of a turbulent jet are usually defined by the exit Reynolds number $Re \equiv U_0d/\nu$ (where $U_0$ is the exit bulk velocity, $d$ is the nozzle exit diameter, and $\nu$ is the kinematic viscosity), the exit radial profiles of mean velocity and turbulence intensity, and the global density ratio of the original-jet fluid ($\rho_a$) to ambient fluid ($\rho_0$), i.e., $R_p \equiv \rho_a/\rho_0$. 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 $R_p \approx 1.0$. 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 $R_p$ 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 $R_p$ do not influence the far-field statistical behavior of the jet. Richards and Pitts\(^16\) later extended the investigation of Pitts’s\(^14,15\) by varying both $R_p$ 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 \(R_e\).

Apparently, there is some conflict 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\(^{14,15}\) 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 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 \(\alpha\) for the unmixedness between the ejected and entrained fluids in a nonreactive turbulent jet.
2. Obtaining the \(\alpha\) 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 \(\alpha\) on the initial conditions of the jet flow.

II. DERIVING THE EXPRESSION FOR \(\alpha = \frac{c_A c_B}{C_A C_B}\) IN A NONREACTING JET

Let \(C_A\) and \(C_B\) represent the instantaneous mass concentrations of the ejected original “warm” fluid of temperature \(\Theta_A\) and the entrained ambient “cold” fluid of \(\Theta_B\) in a turbulent nonreactive jet. Also, let \(\Theta\) 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

\[
C_A + C_B = 1
\]

and

\[
C_A \Theta_A + C_B \Theta_B = \Theta.
\]

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

\[
C_A = \frac{\Theta - \Theta_B}{\Theta - \Theta_A}, \quad C_B = \frac{\Theta_B - \Theta}{\Theta - \Theta_A}.
\]

Then, their fluctuating components are

\[
c_A = \frac{\theta}{\Theta - \Theta_A}, \quad c_B = -\frac{\theta}{\Theta - \Theta_A},
\]

where \(\theta\) is the fluctuating temperature. It follows that the segregation parameter between the ejected and entrained fluids defined by \(\alpha = \frac{c_A c_B}{C_A C_B}\) can be attained from the temperature data, viz.: \(\alpha = \frac{-\theta}{(\Theta - \Theta_B)(\Theta - \Theta_A)}\).

If we, like all measured temperatures reported in the literature, use the relative mean temperatures above the ambient temperature (\(\Theta_0\)), i.e., \(\Theta_B = (\Theta - \Theta_0)\) and \(\Theta_A = (\Theta_B - \Theta_A)\), then after several manipulations, Eq. (6) can be rewritten as

\[
\alpha = -\frac{\theta'}{1 - \alpha},
\]

where \(\theta' = \Theta_{1/2}\) is the RMS temperature. For non-temperature scalars in jets, a similar relation to Eq. (7) for \(\alpha\) 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

\[
\alpha = -\frac{c_A^2}{1 - C_A}
\]

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 \(\alpha\). 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 \(C_A C_B = C_A C_B\) and \(C_A C_B = 0\). 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 \(C_A C_B = C_A C_B + C_B C_B = C_B C_B(1 + \alpha) = 0\) 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 \(C_A C_B > 0\) and \(C_A C_B < 0\). 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, \(\alpha\) 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. deduced that \(\alpha\) should be positive for some cases although being negative for most reactive cases. In addition, for the nonreacting multi-scale mixing or the mixing of two species A and B advected by a single turbulent flow (e.g., jet), it is equally likely for \(\alpha\) to be positive (though mainly negative) because the inherent relations \(C_A + C_B = 1\) and \(C_A + C_B = 0\) for the two-scale 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. 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 \(\alpha\) from Eq. (7), we choose the mean and RMS temperatures (\(\Theta\) and \(\theta'\)) 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 $x$ in the circular jet is confirmed, and so Eq. (7) is validated, by combustion experiments of Langman et al.\cite{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., $x$, $\Theta_{sc}/\Theta_{so}$, and $\theta'/\Theta_{sc}$) 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 $\eta = (x_{LP} - x_{SC})/x_{SC}$ to show more clearly the difference in $x$ between the LP and SC jets. Both jets were measured at $Re = 16 000$. Note also that the data of $\Theta_{sc}/\Theta_{so}$ and $\theta'/\Theta_{sc}$ are reproduced from Mi et al.\cite{12} It is clearly demonstrated that the mean temperature $\Theta_{sc}$ 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 $x$ of the two jets, estimated from Eq. (7) and the data of $\Theta_{sc}/\Theta_{so}$ and $\theta'/\Theta_{sc}$. It is observed that the centerline $x$ is always closer to zero in the LP jet than the SC jet. This difference is demonstrated more obviously by the centerline $\eta$ curve. Interestingly, $\eta$ 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 $\eta$ 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 $(-x_{LP})$ is smaller than the SC-jet one $(-x_{SC})$ 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.

Figure 2 shows the radial profiles of $\eta$, $\Theta_{sc}/\Theta_{so}$, and $\theta'/\Theta_{sc}$ vs $r/(x-x_{o})$, 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 $-x$ and the normalized RMS fluctuation $\theta'/\Theta_{sc}$ across the far-field LP jet are generally smaller than those of the SC counterpart. In particular, the radial profile of $\eta = (x_{LP} - x_{SC})/x_{SC}$ indicates that the unmixedness is substantially greater across the SC jet than the LP jet. On average, the value of $-x$ for the SC jet is about 36% higher than that of the LP jet; note that the averaging is taken over the range of $0 < r/(x-x_{o}) < 0.16$.

Based on the above differences in $x$ and $\theta'/\Theta_{sc}$ 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.\cite{18} 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,\cite{14} 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.\cite{18}) 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 $-x$ and $\theta'/\Theta_{sc}$ are smaller in the LP jet than in the SC jet. This seems to support the conventional view that the magnitude of $\theta'/\Theta_{sc}$ 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.\cite{18} using natural-gas jet

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**FIG. 1.** Variations of the segregation parameter $x$ (——, the blue square) and its difference $\eta$ between the LP and SC jets (——), the normalized mean temperature $\Theta_{sc}/\Theta_{so}$ (——, the blue circle), and the normalized RMS fluctuation $\theta'/\Theta_{sc}$ (——, the brown triangle) in the SC and LP jets both for $Re = 16000$.\textsuperscript{1}\textsuperscript{1} Note: the logarithm is taken on $x$ and $\eta$.

**FIG. 2.** Radial profiles of the segregation parameter $x$ (——, the blue square) and its relative difference $\eta$ between the LP and SC jets (——), the relative mean temperature $\Theta_{sc}/\Theta_{so}$ (——, the blue circle), and the relative RMS fluctuation $\theta'/\Theta_{sc}$ (——, the brown triangle) in the far-field SC and LP jets both for $Re = 16000$.\textsuperscript{1}\textsuperscript{1} Note: the logarithm is taken on $x$ and $\eta$.\textsuperscript{1}
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 ($\chi_r$) of the LP flame against $U_o$ (exacting 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 $Q_f$ being the total flame radiated power (kW) and $Q_r$ the input power (kW). Note also that all the data are reproduced from Figs. 3 and 7 of Langman et al. It is evident that, as $U_o$ rises, $L_f$ gradually increases and $\chi_r$ decreases for both SC and LP flames at $U_o \leq 46$ m/s. When $U_o > 46$ m/s, both $L_f$ and $\chi_r$ 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 $\chi_r$ 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 $x$ 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 $-\chi$ 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 $\chi$. Figure 4 illustrates the radial profiles of $\chi$ obtained in the LP jet ($x/d = 15–65$) and SC jet ($x/d = 8–60$) both for $Re = 16000$. 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 $\Theta_r/\Theta_e$ and $\theta'/\Theta_e$, the radial profiles of $\chi$ 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 $\chi$ 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

$$\chi = -\left(\frac{\Theta_r}{\Theta_e}\right)^2 \left(\frac{\Theta_{ra}}{\Theta_{re}}\right) \left(\frac{\Theta_{re}}{\Theta_{ra}}\right)^{-1}.$$  

(9)

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

$$\chi(\eta) = -g^2(\eta)f^{-1}(\eta) \left[\frac{K(x-x_c)}{d} - f(\eta)\right]^{-1},$$  

(10)

where $f(\eta)$ and $g(\eta)$ are the self-similar functions with $\eta = r(x-x_c)$ and $x_c$ being the $x$-location of the virtual origin; $K$ is a constant. Equation (10) indicates that the radial profiles of $\chi$ 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

$$\chi(x) = -g^2(0)\left[\frac{K(x-x_c)}{d} - 1\right]^{-1},$$  

(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 $\log(-\chi)$ 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 $\chi$, $\Theta_{ra}/\Theta_{re}$ and $\theta'/\Theta_e$ for the circular SC jet ($Re = 16000$) with those for a planar SC jet ($Re = 7620$) studied by Browne et al. 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 $\theta'/\Theta_e$ approaches asymptotically to a far-field value being considerably higher in the circular than in the planar jet. Moreover, $\chi$ is always close to zero for the circular jet than for the planar counterpart. This contrasts strikingly to a higher asymptotic value of $g(\eta = 0)$ or $\theta'/\Theta_e$ 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 $\theta'/\Theta_e$ in the planar jet is not related to a smaller unmixedness of this jet against the circular jet. Moreover, unlike that of $\chi$, the magnitude of $\theta'/\Theta_e$ does not appropriately measure the unmixedness, which was often claimed previously, e.g., by Pitts.

In addition, there is a self-evident finding to be made from Figs. 1 and 5. Namely, an unconfined jet flow can never achieve $\chi = 0$ 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 insight from Eq. (10).
When the jet develops downstream into the self-similar region in the far field, $\theta^*/\Theta_0$ (and, thus, $\theta^*/\Theta_\infty$) will become approximately constant, see Fig. 5. However, the case of $x = 0$ occurs only when $x \rightarrow \infty$, which is practically impossible.

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

As noted in Introduction, Bilger et al.\textsuperscript{10} claimed that the segregation parameter ($\alpha$) 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\textsuperscript{10} and Cai et al.\textsuperscript{11} for a nonpremixed and nonreacting turbulent jet when judging based only on their $x$ measurements. Tong and Warhaf\textsuperscript{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 $\mu$m to measure the fluctuating temperature. Of note, they employed an indirect inference method to obtain $C_2/C_0$ and, thus, $\alpha \equiv C_2/C_0 C_A C_B$. They obtained that $\alpha = -0.04$ to 1.35 and also that the centerline $\alpha \rightarrow -0.04$ at $x/d > 15$. Actually, the turbulent mixing of Tong and Warhaf\textsuperscript{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.

Sixteen years later, Tong's group, i.e., Cai et al.\textsuperscript{11} conducted another investigation on the three-scalar mixing of turbulent jets. Namely, they studied the turbulent mixing between a center acetonedoped 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 $\alpha_0 \equiv \sqrt{C_2/C_1}$. It was found that the centerline $\alpha_0$ 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\textsuperscript{10} and Cai et al.\textsuperscript{11} for their mixing cases in a nonreacting turbulent jet.

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

\begin{align}
\langle \text{Instantaneous} \rangle & \quad C_1 + C_2 + C_3 = 1, \quad (12a) \\
\langle \text{Averaging} \rangle & \quad \overline{C_1} + \overline{C_2} + \overline{C_3} = 1, \quad (12b) \\
\langle \text{Fluctuating} \rangle & \quad \overline{c_1^2} + \overline{c_2^2} + \overline{c_3^2} = 0. \quad (12c)
\end{align}

A couple of manipulations on Eq. (12c) can lead to $\overline{c_1^2} + \overline{c_2^2} = \overline{c_1^2} - \overline{c_1^2}$ and, thus,
whereas earlier than the coflowing air.

Equation (13) implies that $\frac{C_{1}C_{2}}{C_{0}C_{1}C_{2}}$ will become positive as $x$ is sufficiently large for the mixing cases of Tong and Warhaft and Cai et al. 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, $(a_{1}C_{1} + a_{2}C_{2})$ decreases and becomes smaller than $C_{2}$ sufficiently downstream, hence leading to $z_{0} \equiv C_{1}C_{2}/C_{0}C_{1}C_{2} > 0$. This, as demonstrated in Fig. 6, approves the appropriateness of the positive results of Tong and Warhaft and Cai et al.

In fact, it is also reasonable to treat the downstream case of Cai et al. 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

$$z_{1} = \frac{a_{1}C_{1} + a_{2}C_{2}}{C_{0}C_{1}C_{2}} = -\frac{1}{2} \left[ \frac{C_{1} + C_{2}}{C_{1} - C_{0}} \right]^{\frac{2}{3}} \left[ \frac{(C_{1} + C_{2})^{2}}{C_{1}C_{2} - C_{0}C_{1}C_{2}} \right],$$

which is always negative. Note that Eq. (14) is identical with Eq. (8). Based on Eq. (14), $z_{1}$ can be estimated using the data of Cai et al. shown in their Figs. 4 and 5. Figure 6 displays the centerline results of $z_{0}$ and $z_{1}$ vs the data of $x$ shown in Figs. 1 and 5 for the slightly heated SC jet of Mi et al. In addition, we may obtain $z_{2} = -\frac{C_{1}^{2}}{C_{1}C_{2} - C_{0}C_{1}C_{2}}$ [i.e., Eq. (9)] by considering the downstream case of Cai et al. 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., $z_{1}$ and $z_{2}$ become nearly identical at $x/d > 16$, whereas $z_{1} < z_{2}$ 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 $z_{1}$ and $z_{2} \to 0$. 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. than in that of Cai et al. 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. issued into a still surrounding (to be called perhaps “zero-speed coflow”), the one of Cai et al. was accompanied with a low-speed (0.4 m/s) coflow of air. Numerous previous studies, e.g., Refs. 29–31, have demonstrated a very strong effect of the coflow speed on the jet mixing and flame stability. For example, Dahm and Dibble found that an increase of just 1% coflow velocity could result in a 50% reduction in the flame blow-out velocity. Likewise, Han and Mungell 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 $z$. 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 $x$ on both the Reynolds number $Re$ and the density ratio $q$. The use of Eq. (8) enables the calculation of $x$ from the concentration measurements of Pitts with and Dowling and Dimotakis. Figure 7 illustrates the centerline variations of $x$ for different $Re$ jets of C$_3$H$_8$/air by Pitts with $R_p = 1.55$ and those of C$_2$H$_4$/N$_2$ and C$_3$H$_6$/argon by Dowling and Dimotakis with $R_p \approx 1.0$. 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 $z$, obtained from Pitts, is discernible from $Re = 3960$ to $Re = 7930$ but becomes negligible at higher $Re$.

Based on the data of Dowling and Dimotakis, the Reynolds number effect is obviously negligible at $Re > 5000$.
In the present work, we have for the first time derived the expression (7) for the parameter $\alpha$ 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 $C_A C_B = C_A C_B (1 + \alpha) > 0$ and $\frac{c_A}{C_A} = \frac{c_B}{C_B} < 0$ are valid; hence, $-1 < \alpha < 0$. However, for the multiple (>2) scalar mixing, the parameter $\alpha$ cannot always (but mainly) remain negative because $c_A + c_B \neq 0$. Importantly, the magnitude of $\alpha$ reflects the extent of molecular unmixedness between A and B.

The parameter $\alpha$ is related by Eq. (7) or (8) to the scalar mean concentration ($\overline{C}$) and RMS ($\overline{\sigma}$), i.e., the conventional "unmixedness," to represent the real unmixedness between the original-ejected and entrained-ambient fluids.

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 $\alpha$ 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 $\alpha$ is not very significant, the density ratio of jet-to-ambient fluids plays an important role in affecting $\alpha$. Besides, the geometric variation of nozzle configuration strongly influences the magnitude of $\alpha$ over the entire jet flow field.

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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).

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**DATA AVAILABILITY**

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

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