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Effect of Composition-Dependent Viscosity of Liquids on the Performance of Micro-Mixers

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The process of laminar mixing in a T-shaped micro-device is studied by direct numerical simulation for a model binary mixture, composed of two liquids having the same density and the same viscosity, yet presenting a strong fluidity of mixing effect, i.e. the viscosity of the mixture is a function of its composition. In particular, we consider the case where the viscosity of the mixture is up to three times larger than that of the pure liquids, with the maximum viscosity corresponding to either a 50 %-50 % (type 1 mixture), or a 25 %-75 % composition (type 2 mixture), to better emulate the behavior of real mixtures. The results are compared to the case with no fluidity of mixing effects (type 0 mixture), which has been largely investigated previously. In this latter case, the inlet streams remain separated up to a critical Reynolds number, corresponding to a strong increase of the degree of mixing. This transition is also characterized by a symmetry breaking, from a vortex flow regime, with a double mirror symmetry, to an engulfment flow regime, with a point central symmetry. When the fluid mixture has a larger viscosity than that of its pure components, a viscous layer forms at the confluence of the inlet flows, which tends to keep the two streams separated. Therefore, in this case, one would expect that the onset of the engulfment regime should be shifted to larger Reynolds numbers, in comparison with type 0 mixtures, with no sudden increase of the degree of mixing. Although this is what happens for symmetric, type 1 mixtures, for type 2 mixtures we unexpectedly find that, due to the lack of symmetry of the mixture rheology, the transition from vortex to engulfment regime, although occurring at larger *Re*, occurs suddenly, corresponding to a sharp increase of the degree of mixing.

1. Introduction

Efficient mixing of liquids in small volumes is a key target in many processes, such as in miniature fuel cells, molecular diagnostics and, in general, micro-reactors. Among micro-devices, passive micro-mixers are very appealing as they ensure mixing without the help of any external power source. The simplest passive micro-mixer is T-shaped and it is also suitable for fundamental studies, being a junction element in more complex micro-systems. The efficiency of T-shaped micro-mixers for liquid mixing has been largely investigated in the literature and different flow regimes have been identified, both numerically (e.g., Bothe et al., 2006) and experimentally, using water as working fluid (e.g. μ-LIF and μ-PIV experiments by Hoffmann et al., 2006, and flow visualizations by Engler et al., 2004). It was shown that the inlet streams remain separated up to a critical Reynolds number, $Re = 140$, corresponding to the transition from a vortex flow regime, with a double mirror symmetry, to an engulfment flow regime, with a point central symmetry. Then, at $Re = 240$, the flow becomes time periodic, while the transition to chaotic, turbulent regime occurs at $Re > 500$ (Dreher et al., 2009). Fani et al. (2014) applied 3-D linear stability analysis to characterize the instability leading to the time periodic regime, whereas Andreussi et al. (2015) investigated the effect of micro-mixer aspect ratio on flow regimes.

These results have been generalized in a recent work by Galletti et al. (2015), who studied the process of laminar mixing in a T-shaped micro-device by direct numerical simulation for a model binary mixture, composed of two fluids having the same density and the same viscosity, yet presenting a strong fluidity of mixing effect, i.e., the viscosity of the mixture is a function of its composition. In all cases, it was found that the inlet streams remain separated up to a critical *Re*, corresponding to the transition from a vortex to an engulfment flow regime. In the case of a positive fluidity of mixing, the onset of the engulfment regime is

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accompanied by a sharp increase of the degree of mixing, with the critical *Re* decreasing as the fluidity of mixing increases. On the contrary, when the fluid mixture has a larger viscosity than that of its pure components, a viscous layer forms at the confluence of the inlet flows, which tends to keep the two streams separated. This result is in agreement with previous findings by Orsi et al. (2013a), who simulated the mixing process that follows the confluence of two fluids onto a T-junction, comparing the case where the two inlet fluids are both water with the case where one inlet fluid is water and the other is ethanol. Orsi et al. observed that the vortex-engulfment flow transition occurs at larger *Re* numbers for the water-ethanol case than for water-water case (i.e., $Re = 230$ vs. $Re = 140$). The reason of this mixing hindrance was ascribed to the fact that a water-ethanol mixture has a viscosity that is almost three times larger than that of water, so that at the confluence of the T-mixer, the two streams are kept separated from each other by a viscous interfacial layer that hampers vortex formation and retards mixing (Orsi et al., 2013b). In this work, we want to corroborate our previous finding that the main cause of the mixing reduction is the viscosity increase upon mixing; in addition, we also intend to investigate the dependence of the mixing process on the shape of the viscosity-composition diagram $\mu=\mu(\phi)$, namely its skewness.

2. Test case and numerical model

In order to investigate the effect of a composition-dependent viscosity on mixing, we assume that the two fluids composing the mixture have the same density, ρ, and the same viscosity, μ_0, and that, upon mixing, density remains constant, while viscosity increases up to a maximum value, $\mu_{max} = \alpha\mu_0$,. Two types of mixtures are considered in addition to the base case (type 0 mixture) for which $\mu(\phi) = \mu_0$. The first case (type 1 mixture) corresponds to the perfectly symmetric mixture considered in Galletti et al. (2015), with viscosity presenting a simple quadratic dependence on composition,

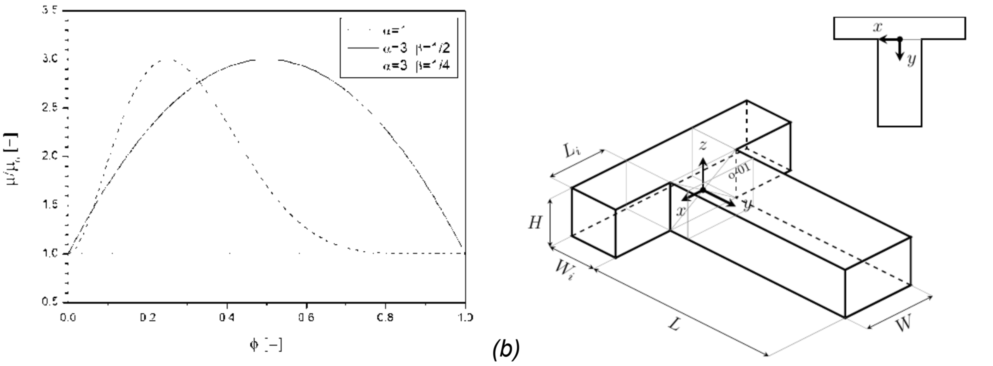
$$\mu(\phi) = \mu_0\left[1 + K_1\phi(1-\phi)\right]; \qquad K_1 = (\alpha-1)\big/\beta^2 \,, \tag{1}$$

where the maximum value, μ_{max}, is reached when $\phi = \beta = ½$, that is for a 50% - 50% mixture. The second case (type 2 mixture) consists of a skewed mixture, whose composition-dependent viscosity,

$$\mu(\phi) = \mu_0\left[1 + K_2\phi^2(1-\phi)^6\right]; \qquad K_2 = (\alpha-1)\Big/\left[\beta^2(1-\beta)^6\right], \tag{2}$$

reaches its maximum value, μ_{max}, at $\phi = ¼$. The viscosity-composition diagram is reported in Figure 1a. The main idea behind these simulations is to investigate the dependence of the degree of mixing on the viscosity gradient. In fact, during the mixing process of a fluid mixture, convective flows are induced by viscosity gradients, due to the non-homogeneous distributions of composition and temperature. Accordingly, we expect that in the skewed case, as the viscosity gradient reaches a larger value than in the symmetric case, the, convective flux will be larger, therefore enhancing the mixing process.

The geometry of the problem consists of the T-shaped micro device shown in Figure 1 along with the reference system. The mixing channel has a rectangular cross section with a 2:1 aspect ratio (i.e. width $W = 2H$ and depth H, so that $d = 4H/3$ is the hydraulic diameter), with length $L = 15d$, while the inlet channels are identical, with square cross section (i.e. width $W_i = H$ and depth H) and length $L_i = 2.25d$. Such length L_i was chosen to include the whole confluence region, while the length of the mixing channel, L, was long enough to completely describe the complex vortical structures and avoid any channel-length effect on the predictions.



(a) (b)

Figure 1 - (a) Viscosity as a function of composition for the three investigated mixtures. (b) Sketch of the T mixer with o-01 section corresponding to y = 1.5d.

Assuming that the heat of mixing and the volume change due to mixing are negligible, so that the process can be modeled to be isothermal and iso-volumetric, at steady state the governing equations are:

$$\rho \mathbf{v}\cdot\nabla\mathbf{v}+\nabla P=\nabla\cdot\left[\mu(\phi)\left(\nabla\mathbf{v}+\nabla\mathbf{v}^{T}\right)\right], \qquad \nabla\cdot\mathbf{v}=0, \qquad (3)$$
$$\mathbf{v}\cdot\nabla\phi=D\nabla^{2}\phi,$$

where **v** is the solenoidal velocity field, P is the modified pressure, ϕ the mass (and volume) fraction of, say, component A of the binary mixture and D is the molecular diffusivity, while the T superscript indicates the transpose of a tensor. If the two fluids are identical, we can imagine adding a very small amount of contaminant, i.e. a dye, to one of the fluids (which therefore continue to have the same physical properties) and therefore, in this case, ϕ indicates the (normalized) dye mass fraction.
Simulations were performed using the commercial software ANSYS Fluent ® v. 15, where dimensions and velocities were scaled in terms of the hydraulic diameter d and the average mean velocity U, i.e. $y^* = y/d$ and $u^* = u/U$.
The grid, obtained from a grid independency study on the velocity field, performed at the highest *Re* number, consists of cubic elements with $H/20$ edge, leading to 20 x 40 elements in each cross section of the mixing channel, thus in agreement with the recommendation by Hussong et al. (2009). A second order discretization scheme was used to solve all the governing equations. Simulations were typically considered converging when the normalized residuals for velocities were stationary with iterations and fell below 10^{-12}. Such small residuals were specifically required to ensure converged solutions near the engulfment (Galletti et al., 2012). The steadiness of the solution with iterations was also assessed by checking the velocity and concentration field in the outlet section of the mixing channels at different iterations.
The boundary conditions consisted of no-slip velocity and no-mass-flux at the channel walls, a constant ambient pressure at the exit, while at the entrance a flow profile corresponding to fully developed flow conditions in a square conduit (Happel and Brenner, 1965) was imposed through a bespoke subroutine. The two fluids were fed with the same flow rate.
Validation of our numerical scheme can be found in Galletti et al. (2015) , where the choice of using the same computational grid to describe both the concentration field and the velocity field is justified for the type 0 mixture. In fact, this is the most critical case, as there is no transversal fluid convection, induced by viscosity gradients, that otherwise prevent the formation of destabilizing sharp concentration gradients. Consequently, as the results for the $\alpha = 1$ case are in perfect agreement with previous investigations, our numerical scheme was successfully validated.

3. Results

In Figure 2 the concentration distributions in the cross-section of the mixing channel located at $y = 1.5\ d$, are shown for different Reynolds numbers in the three cases under investigation.

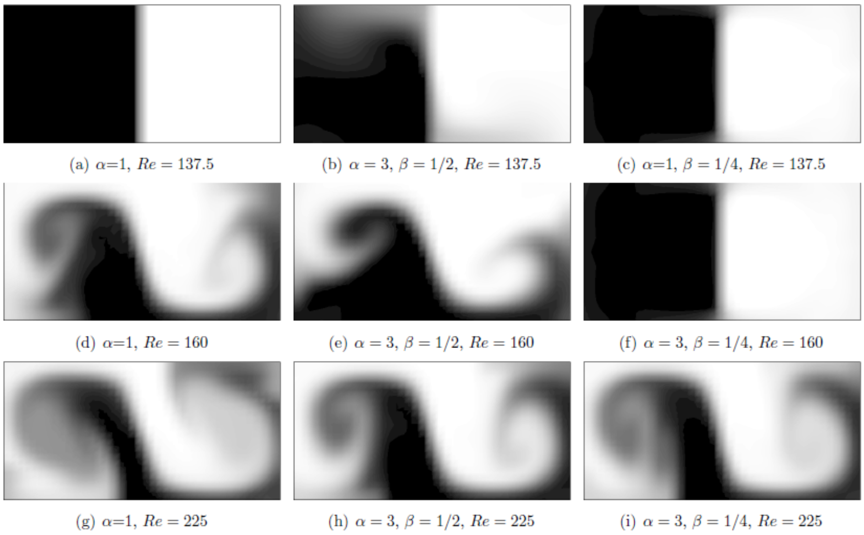
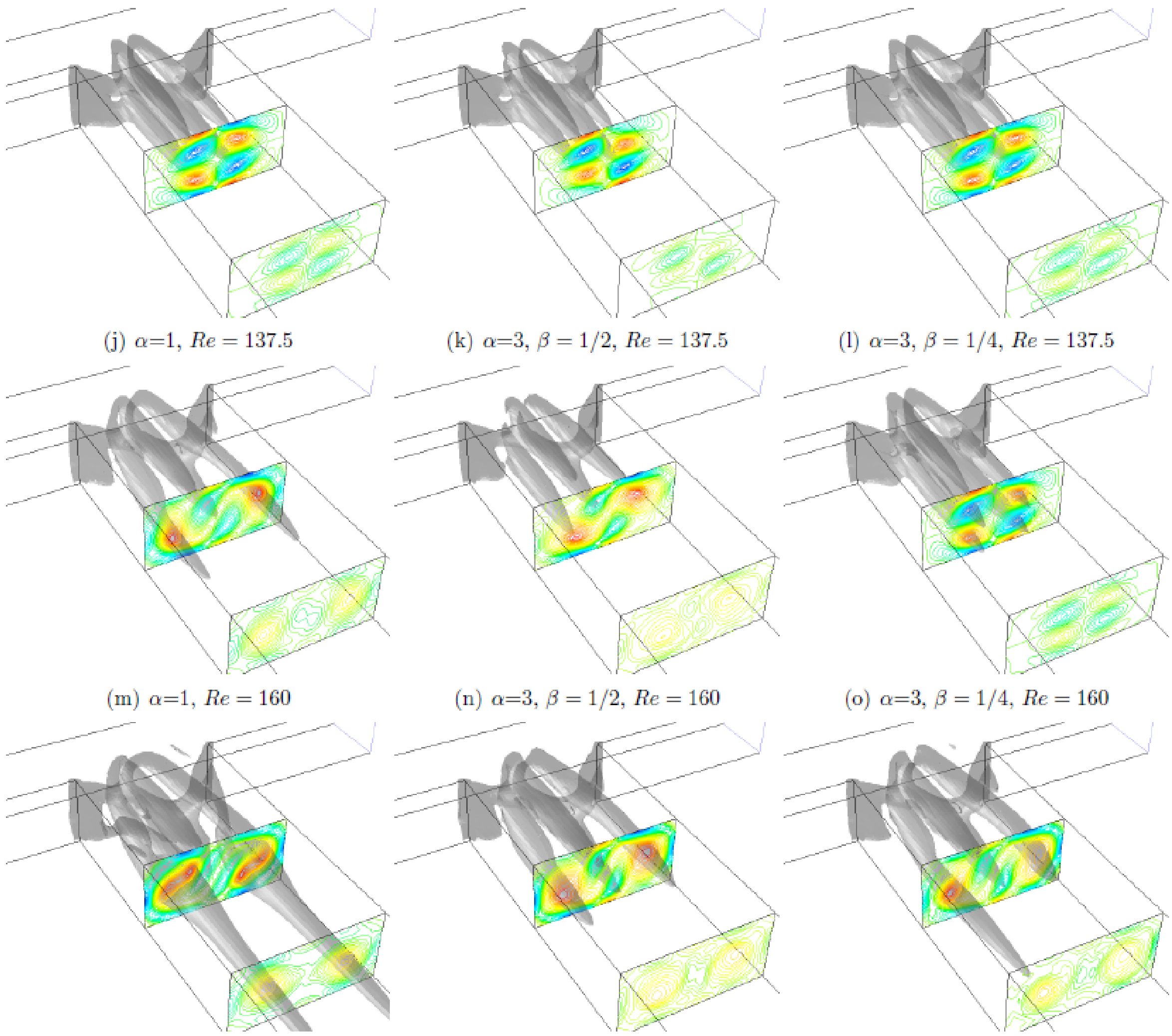


Figure 2: Concentration distribution in the mixing channel cross-section at y = 1.5 d, for type 0 (left), 1 (center) and 2 (right) mixtures, at different Reynolds numbers, i.e. Re = 137.5, Re = 160 and Re = 225.



(j) α=1, Re = 137.5 (k) α=3, β = 1/2, Re = 137.5 (l) α=3, β = 1/4, Re = 137.5

(m) α=1, Re = 160 (n) α=3, β = 1/2, Re = 160 (o) α=3, β = 1/4, Re = 160

(p) α=1, Re = 225 (q) α=3, β = 1/2, Re = 225 (r) α=3, β = 1/4, Re = 225

Figure 3: Identified vortices according to the λ2-criterion and distribution of normal vorticity at y = 1.5 d and y = 3 d, for type 0, 1 and 2 mixtures, at different Reynolds numbers, i.e. Re = 137.5, Re = 160 and Re = 225.

In the same conditions, the vortical structures at the confluence are represented in Figure 3 as identified through the λ_2-criterion (Jeong & Hussain, 1995). Such criterion is based on the concept that a vortex is associated with a local pressure minimum. Accordingly, a vortex is defined as a connected fluid region where the second eigenvalue of the symmetric tensor $S^2+\Omega^2$ is negative, i.e. $\lambda_2<0$, with S and Ω indicating the strain rate and vorticity tensors, respectively, i.e. $\nabla \mathbf{v}=S+\Omega$.

The degree of mixing is evaluated on the basis of volumetric fluxes (see also Galletti et al., 2015) and is defined as:

$$\delta_m = 1-\sigma_b/\sigma_{\max}\,, \tag{4}$$

where

$$\sigma_b = \frac{\int_S \left(\phi(\mathbf{r})-\bar{\phi}_b\right)^2 u(\mathbf{r})\,dxdz}{\int_S u(\mathbf{r})\,dxdz}, \qquad \bar{\phi}_b = \frac{\dot{V}_A}{\dot{V}} = \frac{\int_S \phi(\mathbf{r})\,u(\mathbf{r})\,dxdz}{\int_S u(\mathbf{r})\,dxdz}, \qquad \sigma_{\max} = \sqrt{\bar{\phi}_b\left(1-\bar{\phi}_b\right)}\,. \tag{5}$$

Figure 4 reports the degree of mixing as a function of the *Re* number for the three cases investigated. For the α = 1 base case (type 0 mixture), corresponding to a mixture of two liquids having the same viscosity (and density), the well-known sudden increase of the degree of mixing, occurring at *Re* = 138, due to the onset of the engulfment regime is observed. The transition between vortex and engulfment regimes is particularly evident in Figure 2, where we see that in the vortex regime, when *Re* < 138, the two inlet streams basically remain segregated from each other, while they mix very effectively in the engulfment regime, reaching δ_m =

50% at *Re = 225.* In addition, at *Re* = 138, a symmetry breaking is observed, with a transition from double mirror to central point symmetry structures, where the latter are invariant under point reflection. As for the flow field, the vortical structures of Figure 3 clearly indicate the presence of two stronger legs, extending into the mixing channel, together with other two weaker legs. Such flow structures are very similar to the ones observed by Fani et al. (2013), who used a T-mixer with a different aspect ratio. By further increasing the *Re* number to *Re* = 240, the flow becomes unsteady, showing the presence of periodic motions as indicated in the simulations by Dreher et al. (2009) and also in the stability analysis by Fani et al. (2014).

Type 1 mixtures, that are symmetric and with a negative fluidity of mixing (α = 3), show instead a gradual increase of the degree of mixing, reaching δ_m = 34% when *Re* = 250. Further increasing Re, then, the flow becomes unsteady. In Figure 4 we observe that at *Re* = 125, the degree of mixing is larger than that of the α = 1 base case, because of the positive effect on mixing of the viscosity gradients mentioned above. As *Re* increases, though, the degree of mixing is lower than that of the α = 1 base case, because of the presence of a separating viscous layers, that hampers the development of a strong engulfment flow. It is also worth noting that in the vortex flow regime the degree of mixing is very small for α = 1, while it is different from zero, albeit still small, in the presence of a fluidity of mixing, due to the transversal convective flow induced by viscosity gradients, as it is evident from Figure 2. These observations are confirmed in Figure 3 showing, at lower Re, a double mirror symmetric vortex regime, with similar vortical structures as for the α = 1 case. By increasing Re, the double mirror symmetry disappears, but that happens gradually, as the vortical structures exhibit legs of almost the same length, although the double-mirror symmetry transforms gradually into a central point symmetry. Therefore, we can say that for type 1 mixtures there is no more a sudden transition from the vortex to the engulfment regime, as clearly indicated in Galletti et al. (2015).

Finally, type 2 mixtures, that are asymmetric and with a negative fluidity of mixing (α = 3), exhibit a behavior that is, surprisingly, different from the other two cases. In fact, from Figures 2 and 4 we see that the vortex-engulfment transition occurs suddenly, unlike for type 1 mixtures, and at a larger Reynolds number. In addition, in Figure 3 we see that as the Re number increases, the double mirror symmetry disappears completely, as the vortical structures exhibit two legs of different lengths. Therefore, the asymmetry introduced by the non-symmetric viscosity-concentration dependence of Figure 1b, does not allow the formation of a central point symmetric morphology, thus strengthening the engulfment regime and enhancing the mixing process. This effect is further increased by the transversal convection induced by the viscosity gradients that, compared to the ones that have been observed in type 1 mixtures, are larger, due to the steeper viscosity-concentration curve. Moreover, the occurrence of the unsteady flow is anticipated at *Re* = 225.

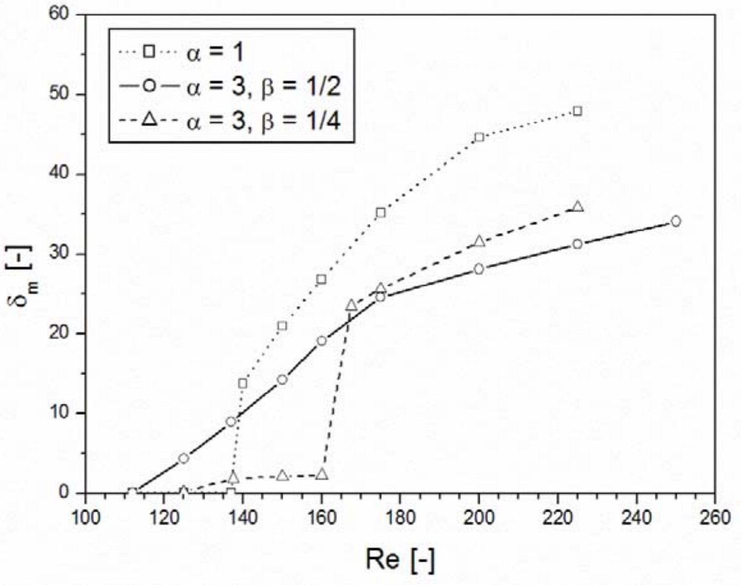


Figure 4: degree of mixing at the mixing channel outlet section , i.e. y = 15 d, for the three type mixtures.

4. Conclusions

Laminar mixing of non-regular binary mixtures is studied numerically, considering the effect on the velocity field of the viscosity change of the two fluids as they are brought in contact with one another. A model mixture is considered, consisting of two fluids having the same density and the same viscosity, μ_0, yet presenting a strong fluidity of mixing effect, i.e., the viscosity of the mixture is a function of its composition. In particular, we considered three cases, where either $\mu=\mu_0$ (this is the base case, indicated as the type 0 mixture) or μ is larger than μ_0, reaching a maximum value, $\mu_{max}=3\mu_0$, either at $\phi=½$ (symmetric, type 1 mixture), or at $\phi=¼$ (asymmetric, type 2 mixture).
In the base case, we observe that the flow field undergoes a sudden transition from a mirror-symmetric vortex regime to a point-symmetric engulfment regime at *Re* = 138, with a sharp increase of the degree of mixing. On the other hand, with type 1 mixtures, the degree of mixing is non zero, albeit very small, even at smaller *Re*, but then the engulfment regime is reached at larger *Re*, without any sharp transition from vortex to engulfment. Finally, with type 2 mixtures, the transition occurs suddenly, but at larger *Re = 160*. The explanation of this unexpected behavior is that the asymmetry of the type 2 mixture rheology prevents the flow field from assuming any symmetric morphology, thus accelerating the mixing efficiency of the engulfment regime.

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