

Signature of coalescence during scalar mixing in heterogeneous flow fields

Sabyasachi Sen¹, Prajwal¹, Joris Heyman², Tanguy Le Borgne², Aditya Bandopadhyay¹

¹Department of Mechanical Engineering, Indian Institute of Technology Kharagpur, West Bengal - 721302, India

²Geosciences Rennes, UMR 6118, Université de Rennes 1, CNRS, 35000 Rennes, France



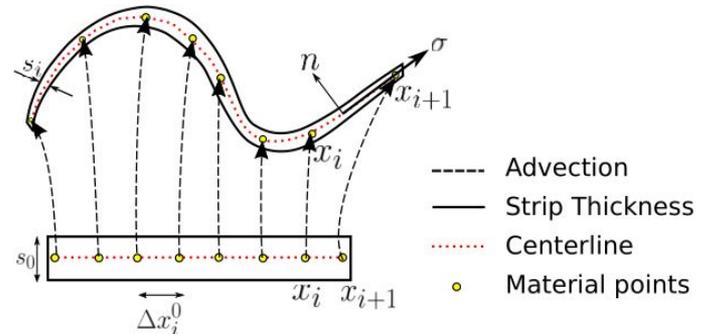
Motivation

- The influence of mixing on reaction kinetics has been studied for the mixing of waters with different compositions
- Recently, emphasis has been put on the role of fluid deformation on mixing rates and its consequences for reactivity
- However, predicting mixing rates and understanding their influence on reaction kinetics remains a challenge particularly when there is a reconnection between several parts of the mixing interface, leading, at large mixing time, to a so-called coalescence regime
- Understanding the effect of this coalescence regime becomes important in flows of geophysical relevance since many biogeochemical processes have been shown to be controlled by mixing

Aim of this presentation

- We study the effect of stretching-enhanced mixing and coalescence on reactivity through scalar transport in a Rankine vortex (where material lines elongate linearly in time) and in a Stokes flow through a 3D bead pack (where material lines elongate exponentially in time)
- We employ a lamellar approach to solve for the transport problem which allows us to gain insight into coalescence dynamics
- Analytical solution for solute concentrations is substituted in the speciation equation for a mixing-limited, heterogeneous reaction to obtain closed form solutions for the upscaled reactivity and mass of precipitate as a function of time
- We compare the integrated reactivity obtained from the independent lamellar model and that from a numerical reconstruction using the diffusive strip method and highlight the impact of coalescence on reaction kinetics

Schematic of tracer representation using a finite number of points at the initial time instant - shown as a straight strip, and at a later time instant - in a deformed configuration



Obtaining the conservative scalar transport equation

The reaction to be studied is given by $A_{(aq.)} + B_{(aq.)} \rightleftharpoons C_{(s)}$

$$\begin{array}{ccc}
 \frac{\partial a}{\partial t} + \mathbf{v} \cdot \nabla a = D \nabla^2 a - r & \begin{array}{c} \text{Under the Ranz} \\ \text{transformation} \\ \frac{d\tau}{dt} = \frac{D}{s^2} \\ \tilde{n} = n/s \end{array} & \frac{\partial a}{\partial \tau} = \frac{\partial^2 a}{\partial \tilde{n}^2} - \frac{s^2}{D} r \\
 \frac{\partial b}{\partial t} + \mathbf{v} \cdot \nabla b = D \nabla^2 b - r & \longrightarrow & \frac{\partial b}{\partial \tau} = \frac{\partial^2 b}{\partial \tilde{n}^2} - \frac{s^2}{D} r \\
 \frac{\partial c}{\partial t} = r & & \begin{array}{c} \text{Using the concept} \\ \text{of components} \\ \text{substituting } q=a-b \end{array} & \frac{\partial q}{\partial \tau} = \frac{\partial^2 q}{\partial \tilde{n}^2}
 \end{array}$$

Here, q represents a conservative scalar and the solution to the reduced governing equation is given by:

$$q(\tilde{n}, t) = \frac{1}{\sqrt{1 + 4\tau(t)}} \exp\left(\frac{-\tilde{n}^2}{1 + 4\tau(t)}\right)$$

This expression is used in our study of the transport of a conservative scalar in a Rankine vortex and in a 3D flow with chaotic streamlines

Reactive transport equations

Under the assumption of a mixing-limited reaction, the chemical relation equation takes the form of the following chemical equilibrium equation

$$ab = K_{eq}$$

In terms of the conservative component q , the concentrations of a and b can be expressed as

$$a = \frac{q + \sqrt{q^2 + 4K_{eq}}}{2}, b = \frac{-q + \sqrt{q^2 + 4K_{eq}}}{2}$$

This allows us to express the rate of product formation in the following form:

$$\frac{\partial c}{\partial \tau} = \frac{s^2}{D} r = \frac{2K_{eq}}{(q^2 + 4K_{eq})^{3/2}} \left(\frac{\partial q}{\partial \tilde{n}} \right)^2$$

This leads to the dimensional form of the reactivity which is a key result since it highlights the explicit dependence of the reactivity on the mixing rate.

$$r = \frac{1}{4Pe\sqrt{K_{eq}}} \left(\frac{\partial q}{\partial n} \right)^2 \text{ in the limit } K_{eq} \gg q^2$$

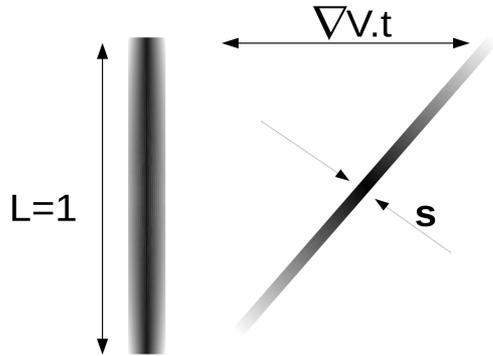
Finally, the mass of precipitate is obtained:

$$m_c = \int_0^\tau \int_{l_{strip}} \int_{-\infty}^{\infty} \frac{\partial c}{\partial \tau} dndld\tau = s_0 l_0 \int_0^\tau \int_{-\infty}^{\infty} \frac{\tilde{n}^2}{\sqrt{K_{eq}}(1+4\tau)^3} \exp\left(\frac{-2\tilde{n}^2}{1+4\tau}\right) d\tilde{n}d\tau = \frac{s_0 l_0}{16} \sqrt{\frac{2\pi}{K_{eq}}} \left(1 - \frac{1}{\sqrt{1+4\tau}}\right)$$

Mixing in a Rankine vortex

$$v_{\theta} = \begin{cases} c_1 r & r \leq r_c \\ \frac{c_2}{r} & r > r_c \end{cases}$$

Rankine vortices are frequently used to model tornadoes and mesocyclones; at the same time, the outer free vortex region of the flow model resembles a point vortex which is a common approximation to circulating regions of stagnant fluid in porous media flows



$$s = \frac{s_0}{\sqrt{1 + \nabla v^2 t^2}}$$

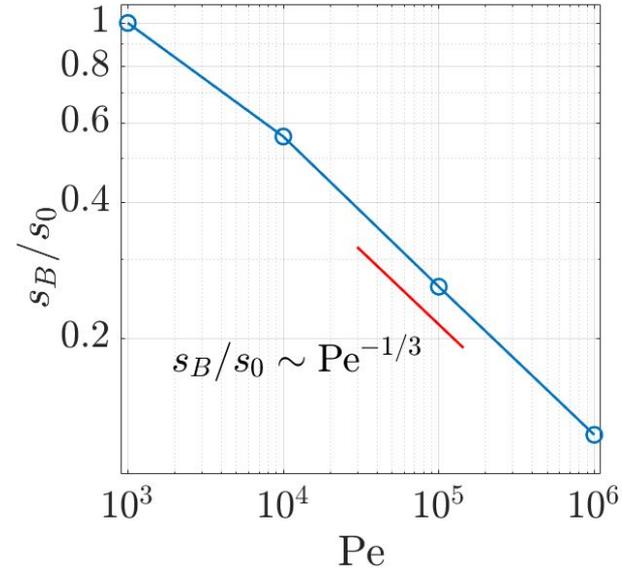
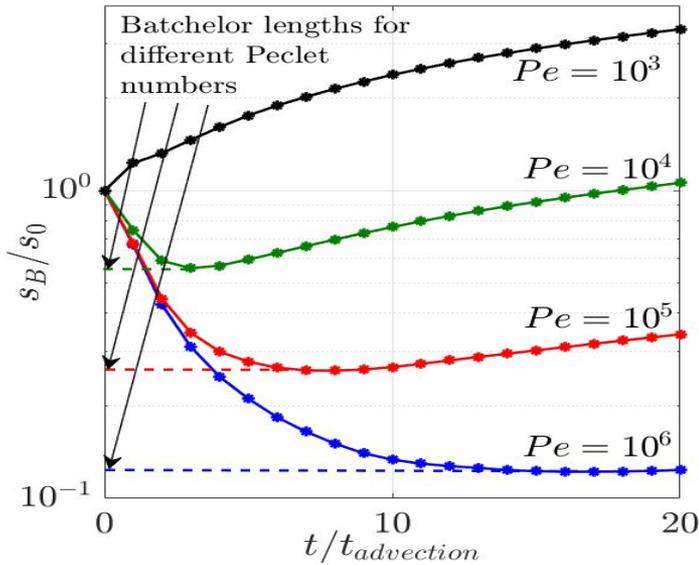
When shear effects are significant

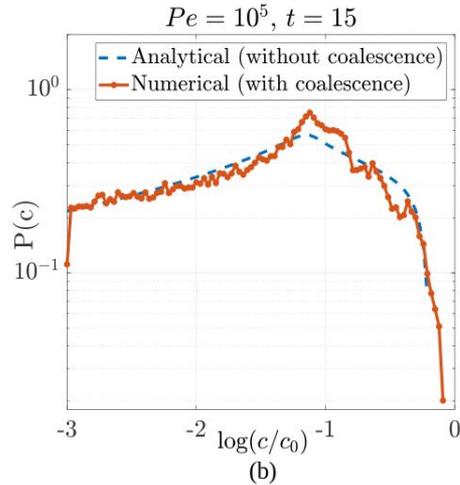
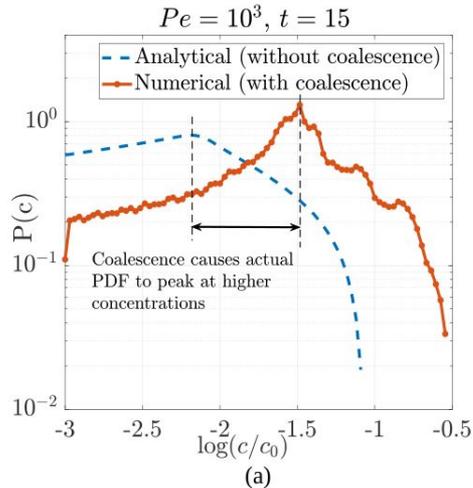
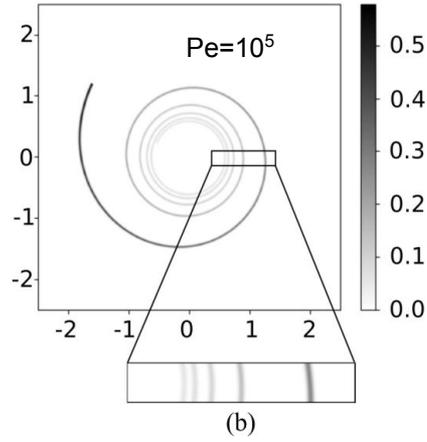
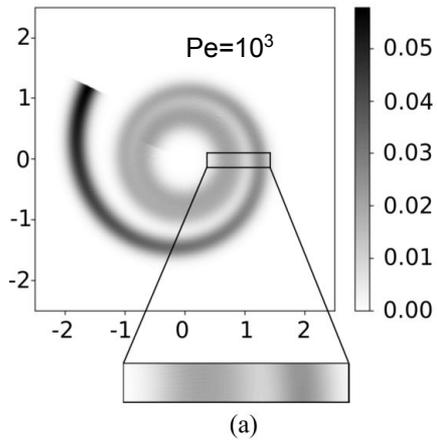
$t_{shear} \approx \frac{1}{\nabla v}$

$$s \approx \frac{s_0}{\nabla v t}$$

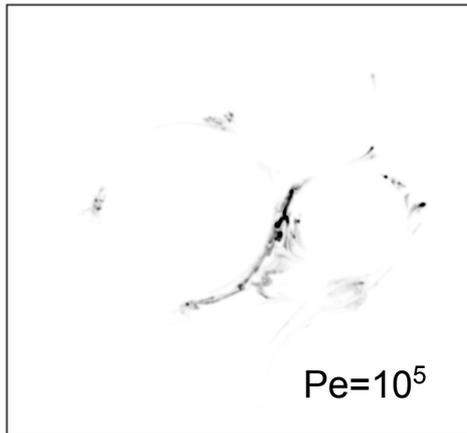
$$\frac{s_0}{\nabla v t} \sim \sqrt{Dt} \quad \Rightarrow \quad t_{mix} \sim \nabla v^{-1} Pe^{1/3}$$

Where $Pe = \frac{t_{diffusion}}{t_{shear}} = \frac{s_0^2 \nabla v}{D}$

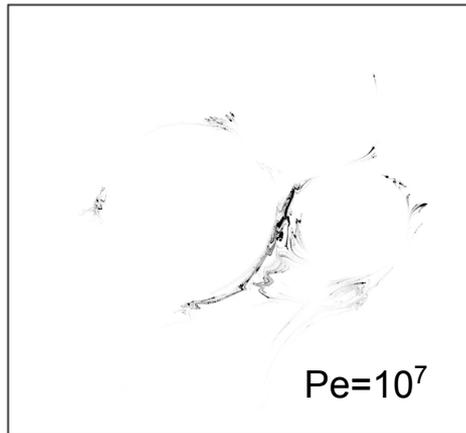




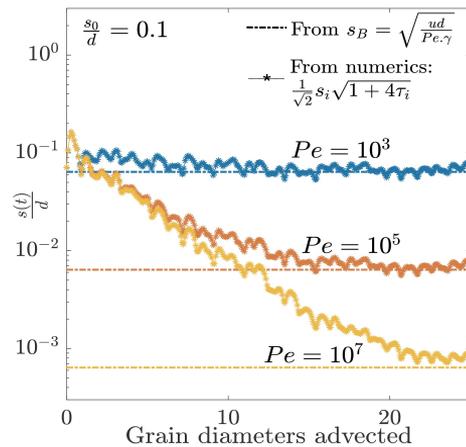
- Reconstruction of scalar for $Pe=10^3$ and $Pe=10^5$ and the probability density function (PDF) of concentration are shown in these figures
- The difference between the numerical PDF and the analytical PDF in intermediate and high concentration zones is essentially how the presence of coalescence manifests itself in the global PDF of concentration.



(a)



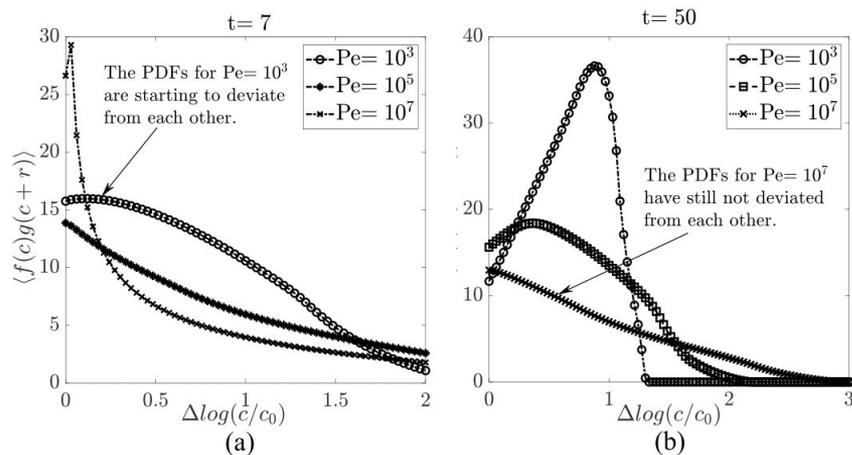
(b)

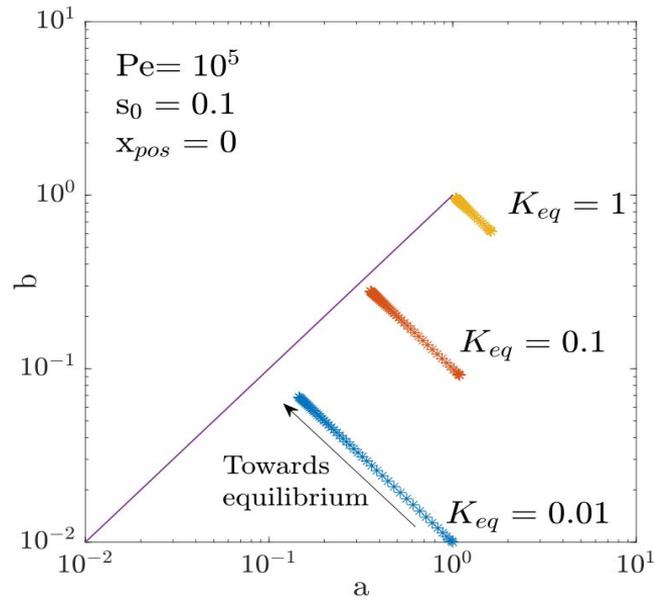


(c)

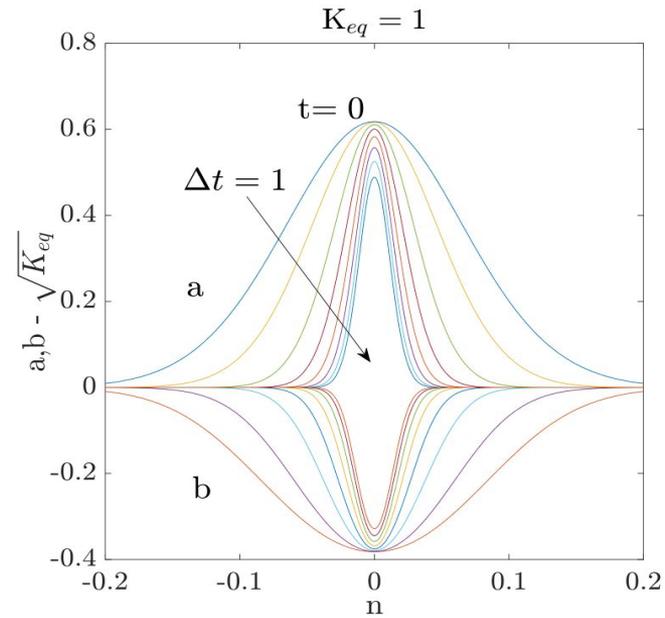
Figures (a) and (b) show the reconstructed concentration fields for two Peclet numbers in a Stokes flow through a periodic bead pack; Figure (c) shows the strip width reaching and staying fixed to the Batchelor length in such a flow - where the material line elongates exponentially in time.

Below: The cross correlation function of the analytical and numerical PDF as a heuristic for the onset of coalescence



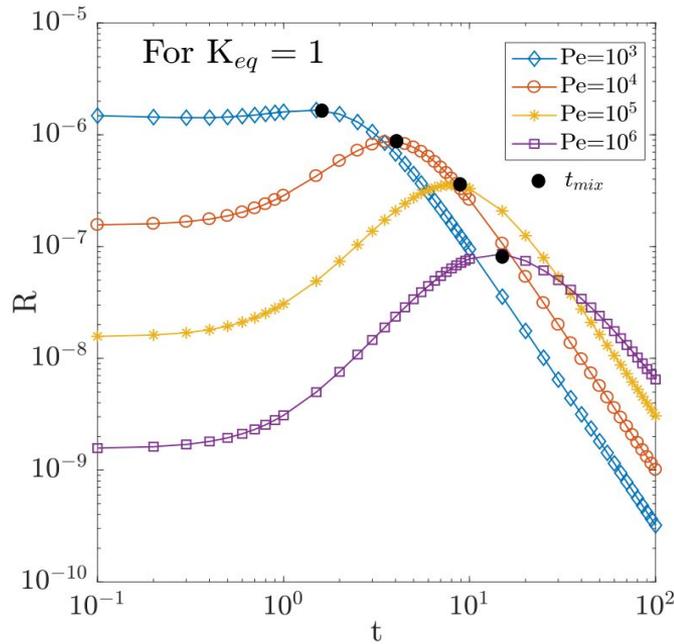


(a)

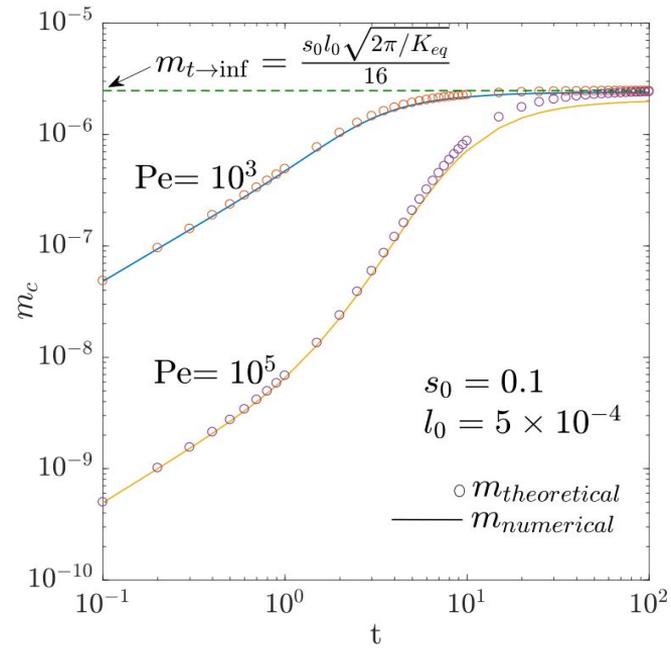


(b)

a) In a linear shear flow, we plot the evolution of the reactant concentration towards the global equilibrium state for different equilibrium constants. From the density of the markers we can see that initially, the concentration a evolves at an increasing rate and then at a decreasing rate as it approaches the equilibrium line. This is an indicator of the driving force for the reaction, which is the reactivity. The faster increments in the concentration of a occur until the mixing time, where the concentration gradients are the highest and in turn, the reactivity is also at its peak, beyond which the approach becomes slower. b) The evolution of a and b towards the equilibrium concentration $\sqrt{K_{eq}}$ in a linear shear flow.

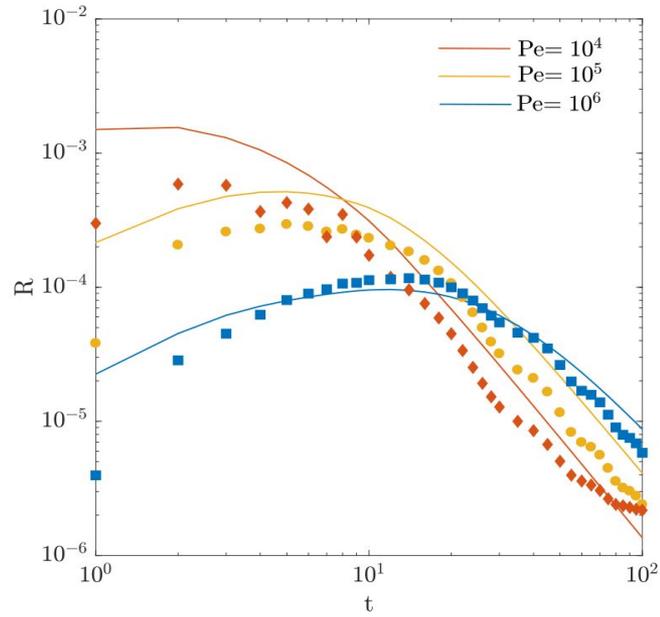


(a)

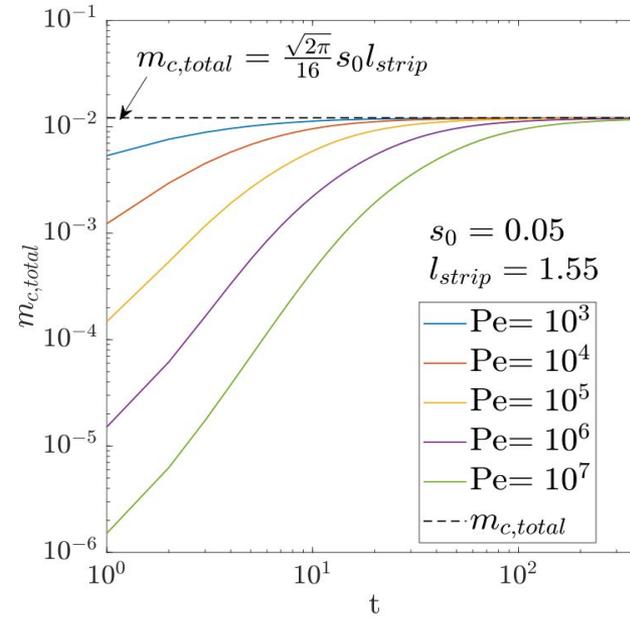


(b)

a) The integrated reactivity $R = \iint r \, dn \, dl$ is plotted against time for a solitary strip in a linear shear flow for a range of Peclet numbers (with $K_{eq}=1$). In each case, the reactivity in the initial stages is constant and is seen to scale as $1/Pe$ and as expected from the lamellar description of mixing, decreases beyond the mixing time indicated by the black markers. b) The total mass obtained in the domain is plotted for various Peclet numbers. All yield the same final mass but do so at different times. Low Peclet number flows reach mixing time before that of high Peclet number flows and react faster to give the same final mass.



(a)



(b)

In this plot, we highlight the effect of coalescence on reactivity for a Rankine vortex. The integrated reactivity $R = \iint r dndl$ from the independent lamella model (solid line) is compared to that obtained from the numerical reconstruction using the diffusive strip method (markers). The reactivity in the presence of coalescence or equivalently, in the absence of discrete lamellar structures is seen to be less than that in the absence of coalescence. The onset of coalescence smothers concentration gradients and this in turn reduces the reactivity. Thus, the role of coalescence is seen to be that of decreasing the rate of product formation.

Conclusion

- A key signature of coalescence during scalar mixing has been identified as the difference between the probability density function (PDF) of concentration, obtained from numerical simulations by exploiting the linear nature of the Fourier equation, and the concentration PDF obtained from the independent lamella model which does not account for the phenomena of strip overlap.
- The role of fluid deformation due to the inherent heterogeneity in the flow is shown to significantly enhance reactivity than that for a homogeneous flow. Concentration gradients and reactivity are observed to increase until the Peclet number dependent mixing time, beyond which gradients are smothered and a simultaneous decay in reactivity occurs.
- The mass of precipitate after sufficient time is shown to be independent of Peclet number and the role of the Peclet number is seen to be important only at $t < t_{\text{mix}}$ for the mass dynamics. Finally, we have highlighted the impact of coalescence, during scalar transport in a Rankine vortex, on reaction kinetics where the smothering of concentration gradients due to the onset of coalescence is shown to decrease the upscaled reactivity in comparison to an independent lamella model

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