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Enhanced reaction kinetics and reactive mixing scale dynamics in mixing fronts under shear flow for arbitrary Damköhler numbers

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Abstract

Mixing fronts, where fluids of different chemical compositions mix with each other, are known to represent hotspots of chemical reaction in hydrological systems. These fronts are typically subjected to velocity gradients, ranging from the pore scale due to no slip boundary conditions at fluid solid interfaces, to the catchment scale due to permeability variations and complex geometry of the Darcy velocity streamlines. A common trait of these processes is that the mixing interface is strained by shear. Depending on the Péclet number Pe , which represents the ratio of the characteristic diffusion time to the characteristic shear time, and the Damköhler number Da , which represents the ratio of the characteristic diffusion time to the characteristic reaction time, the local reaction rates can be strongly impacted by the dynamics of the mixing interface. So far, this impact has been characterized mostly either in kinetics-limited or in mixing-limited conditions, that is, for either low or high Da . Here the coupling of shear flow and chemical reactivity is investigated for arbitrary Damköhler numbers, for a bimolecular reaction and an initial interface with separated reactants. Approximate analytical expressions for the global production rate and reactive mixing scale are derived based on a reactive lamella approach that allows for a general coupling between stretching enhanced mixing and chemical reactions. While for $Pe < Da$, reaction kinetics and stretching effects are decoupled, a scenario which we name "weak stretching", for $Pe > Da$, we uncover a "strong stretching" scenario where new scaling laws emerge from the interplay between reaction kinetics, diffusion, and stretching. The analytical results are validated against numerical simulations. These findings shed light on the effect of flow heterogeneity on the enhancement of chemical reaction and the creation of spatially localized hotspots of reactivity for a broad range of systems ranging from kinetic limited to mixing limited situations.

Keywords: Reactive front, mixing, arbitrary Damköhler number, shear flow, reaction width

1. Introduction

Reaction fronts where two reactive fluids displace one another play an important role in a range of processes, including contaminant plume transport and reaction, soil and aquifer remediation, CO₂ sequestration

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11 and the development of hotspots of reaction in mixing zones [1, 2, 3, 4, 5, 6, 7]. Mixing of reactants by het-
12 erogeneous flows leads to the formation of geometrically-complex fronts, at which chemical reactions occur
13 [8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18], Such fronts are subjected to fluid deformation which increases the sur-
14 face available for diffusive mass transfer thereby enhancing effective reaction rates [19, 20, 21, 22, 23, 24, 25].

15 The dynamics of reactive mixing systems has been widely studied in the absence of velocity gradients.
16 Galfi and Racz [26] and Larralde et al. [27] studied diffusion coupled to the reversible bimolecular reac-
17 tion $A + B \rightleftharpoons C$ for the case of initially well separated reactants at different bulk concentrations. The
18 mathematical insights obtained through the long-time asymptotics are that the mass of product formed,
19 m_c , grows as $m_c \sim t^{1/2}$, which is expected from the diffusive flux across the interface, while the width of
20 the reaction front s_r grows as $s_r \sim t^{1/6}$, due to the balance between the diffusive growth and the reactive
21 consumption. Taitelbaum et al. [28] had studied the initial time dynamics of such reactive fronts for a
22 bimolecular reaction and showed that the rate of production of the product grows as $t^{1/2}$ as opposed to the
23 asymptotic long-time limit t^{-1} . For a similar reactive front, Arshadi and Rajaram [29] found by means of
24 regular perturbation analysis that the growth rate of the total product mass behaves as $dm_c/dt \sim t^{1/2}$ at
25 short times while at larger times it evolves as $dm_c/dt \sim t^{-1/2}$; the transition time between the two regimes is
26 shown to depend on the rate constant and diffusion coefficient. Similar observations of the kinetic diffusive
27 regimes were observed in the work by Chopard et al. [30] who focused on quantification of the influence of
28 the reversible reaction in comparison to the forward reaction, with an emphasis on the formalism of cellu-
29 lar automata. This problem was also addressed through a mean-field formalism for species with different
30 reactivities by Sinder and Pelleg [31] with the product formed being immobile. Results for reactive fronts
31 for the annihilation reaction ($A + B \rightarrow \Phi$) were obtained through probabilistic cellular automata for a 1D
32 case [32] where it was shown that the reaction width grows as $w \sim t^{1/4}$. The validity of the analytical
33 results were then investigated for higher dimensions by Howard and Crowdy [33]. They found that the char-
34 acteristic time of the crossover between the initial time irreversible and the long time reversible behaviour
35 depends on the inverse of the Damköhler number based on the reversible kinetics rate constant. Several
36 other works relate to reaction-diffusion waves in autocatalytic systems ($A + nB \rightarrow (n + 1)B$) [34] where
37 the front propagation velocity is obtained analytically in the long time limit. Merkin et al. [35, 36, 37]
38 also developed solutions for the situations of well stirred mixtures and low catalyst concentrations where
39 the catalyzer decays in time through a parallel reaction ($B \rightarrow C$). Taitelbaum [38] studied the influence
40 of initial conditions and fluctuations on the segregation and patterns formed by such bimolecular systems.
41 Taitelbaum et al. [39] studied the influence of bias on the reaction kinetics of bimolecular reactive fronts.
42 This study was motivated by the observations of Koo and Kopelman [40] that the reaction kinetics of in-
43 jected fronts, i.e. without a stationary front, do not follow the analytical predictions of Galfi and Racz [26].
44 Park et al. [41] performed experiments to prevent the advective motion of such injections and showed that
45 the analytical predictions of the scalings of the reactive fronts agrees well with experimental observations of

46 copper ion-complex formation reactions. Havlin et al. [42] and Bazant and Stone [43] have also considered
47 the scenario of diffusion-reaction kinetics for a system with one static component (for example, a solid porous
48 catalyst). Through analytical solutions, Taitelbaum and Koza [44] have shown that the reaction fronts for
49 initially well separated reactants may move forward or backward depending upon the relative diffusivity of
50 the two species reacting species (see also [45, 46]). Benson et al. [47] have analyzed an annihilation reaction
51 between two reactants to study the mixing density. Sinder and Pelleg [48] analyzed the reactive boundary
52 layer between a two species reversible reaction system ($A \leftrightarrow B$) by means of a singular perturbation analysis
53 for fast reactions, i.e where the characteristic reaction time is much smaller than the characteristic diffusion
54 time. On similar lines, Sinder and Pelleg [49] analyzed a system with two competing reactions and found
55 analogous irreversible initial regimes followed by a crossover regime depending on the reaction kinetics. It
56 is shown that the reversible time regime encompasses two distinct reversible and irreversible zones near the
57 reaction front. When velocity gradients exist in a fluid flow, transported reactive mixtures are submitted to
58 repeated stretching actions that lead to the formation of elongated lamellar structures. The latter are known
59 to promote mixing and enhance reaction rates [19, 50, 51, 52, 53, 54, 55]. This problem was studied by Ranz
60 [56], who showed that the coupling of lamella deformation with diffusion can be reduced to a 1D diffusion
61 reaction-diffusion equation by making use of (i) the rescaled coordinate perpendicular to the direction of
62 lamella elongation and (ii) the so-called warped time that rescales temporal increments with the lamella
63 elongation to eliminate the stretching term. Qualitative insights about the coupling of stretching-enhanced
64 mixing and chemical reactions were obtained in [20, 57, 58] based on numerical simulations. Le Borgne et
65 al. [25] investigated the impact of non-uniform flow conditions on the mixing and reaction rates under such
66 conditions using a lamellar mixing front approach. Paster et al. [59] investigated the impact of shear upon
67 reaction for uniformly initial distribution of reactants with concentration fluctuations.

68 Fluid stretching has been shown to play a fundamental role for governing mixing in porous media
69 [60, 61, 62, 63, 64]. The presence of heterogeneity in the advective flow field invariably leads to lamella
70 formation and subsequent coalescence [65, 66, 67, 68]. Therefore, understanding the interaction between the
71 invading fluid and the residing fluid is imperative towards prediction of species transport in such media. For
72 example, Mays and Neupauer [69] have demonstrated a methodology to achieve enhanced mixing inducing
73 chaotic Darcy-like flow patterns. Gramling et al. [70] performed an experimental and theoretical Darcy
74 scale study of the reaction rate and moving front width in a porous medium for the irreversible bimolecular
75 reaction $A+B \rightarrow C$. They provided hints that incomplete pore-scale mixing was limiting local reaction rates,
76 based on the inability of a Darcy scale modeling approach to properly predict the longitudinal concentration
77 profiles and total mass of product. Later, de Anna et al. [24] investigated the impact of pore-scale mixing
78 on chemical reactions in a two-dimensional (2D) porous medium consisting of cylindrical grains allowing to
79 measure the 2D concentration field at the pore scale. They confirmed the role of mixing in controlling local
80 reaction rates and were able to quantitatively predict the temporal evolution of the measured product mass

81 in the infinite Damköhler limit (i.e for fast reactions) using an upscaling theory based on the concept of
82 lamellar reaction front [71]. Oates [72] and later Chiogna and Bellin [73] used a concentration probability
83 density function (PDF) based mixing model to quantify the observed reaction rates in the experimental
84 setup of Gramling et al. [70].

85 In this work, we focus on reactive mixing of the bimolecular reaction $A + B \rightarrow C$ under shear flow, which
86 represents a fundamental fluid deformation and reaction process in porous media. Shear occurs for any
87 situations where adjacent parallel streamlines have different velocities. At the pore scale, shear is created by
88 strong velocity gradients near fluid-solid interfaces [24]. At the Darcy scale, shear flow is the dominant fluid
89 stretching mechanism governing mixing and spreading in randomly stratified media, typical of sedimentary
90 formations, where the horizontal correlation length is much larger than the vertical one [74, 75, 76, 77, 78].
91 More generally, shear plays an important role in heterogeneous porous media as one of the components
92 of local deformation tensors [79, 66]. While linear shear flows give rise to a linear elongation of mixing
93 fronts, other stretching dynamics could be considered in the same framework, such as power law [66] or
94 exponential [80] deformation (which implies that the length of a material line increases in time following a
95 power-law behaviour in time or increases exponentially). Hence, beyond the particular choice of shear flows,
96 the general methodology presented here for predicting the effective reaction kinetics of mixing fronts from
97 the coupling between the stretching dynamics and the characteristic reaction time scales is likely relevant
98 for more complex flows. While the impact of fluid stretching on reaction kinetics is well understood for
99 fast reactions [81, 25], the broad range of scales and the diversity of chemical reactions entails the need
100 for accounting for a wide range of reaction time scales compared to the transport time scales, that is, for
101 a wide range of Damköhler numbers [82, 83, 84]. Therefore, we investigate analytically and numerically
102 the global reaction kinetics of a mixing front with two initially separated diffusive reactants subjected to
103 velocity gradients of different magnitudes, for an arbitrary finite Damköhler number. In particular we are
104 interested (i) in the rate at which the product is formed and (ii) in the temporal evolution of the width of the
105 reaction front. From the analysis of asymptotic behaviors of pertinent equations we establish the existence of
106 three distinct regimes of effective upscaled reaction kinetics that result from the interplay between reaction,
107 diffusion and stretching. We pinpoint the transition times which demarcate the various regimes. We validate
108 the analytical predictions derived for the temporal scalings of the product mass and the reaction mixing
109 scale, i.e. the width of the reaction front, by comparison with numerical simulations.

110 2. Theoretical framework

111 2.1. System description and governing equations

112 We consider the scenario of two reactants A and B that are initially separated by an interface. The
113 system is subjected to a linear shear flow perpendicular to the initial interface characterized by a constant

114 velocity gradient as shown in figure 1.a. The designated mass of reaction product grows and spreads owing to
 115 (a) diffusion, (b) advection (imposed shear flow) and (c) reaction at the interface between the two reactants.
 116 The rate of reaction is assumed to follow a second order kinetics given by $A + B \xrightarrow{k} C$ with k denoting
 117 the rate constant of the chemical reaction. We consider the following two coordinate systems illustrated
 118 in figure 1.b. The coordinate system (x, y) is fixed while the frame (x', y') is attached to the stretching
 119 interface so that the coordinate x' always corresponds to the direction perpendicular to the interface. The
 120 shear flow in the laboratory frame is denoted by $\tilde{\mathbf{u}} = (G\tilde{y}, 0)$. Owing to the action of the shear, the front
 121 stretches about the origin as the pivot. As stretching occurs along the interfacial direction, the thickness
 122 in the direction normal to the front decreases due to the constraint of continuity (that is, the fluid mass
 123 conservation). In other words, stretching of the fluid along the interface is accompanied by a compression
 124 in the perpendicular direction of the interface.

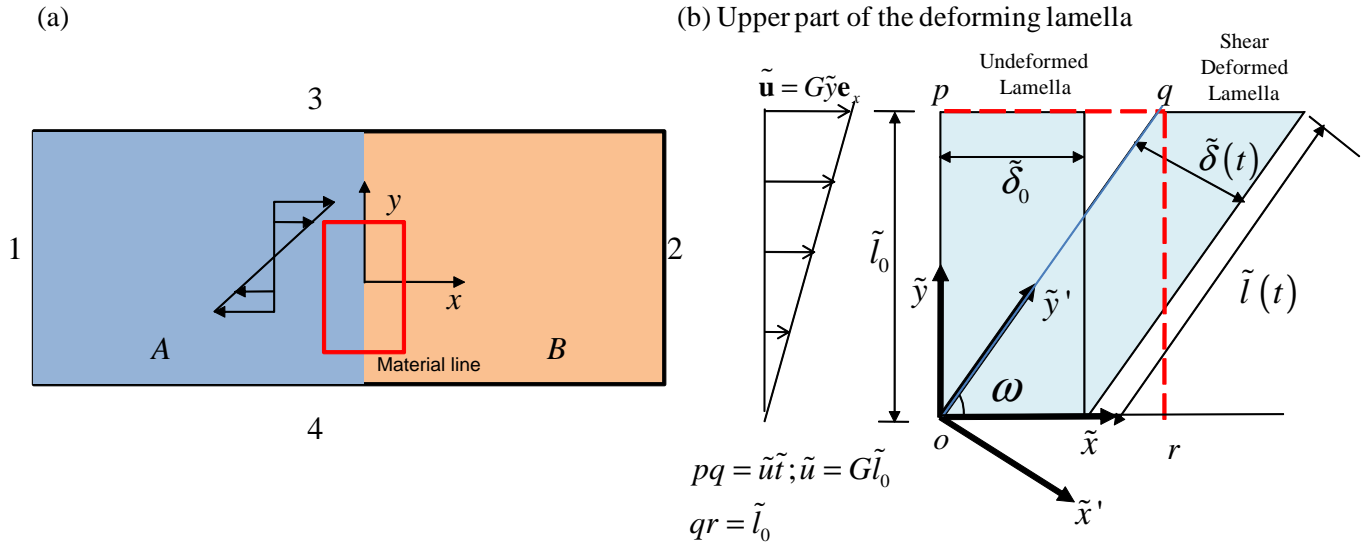


Figure 1: General schematic and description of the system employed for a 2D simulation of reaction-diffusion in the presence of a linear shear flow. (a) Boundaries 1 and 2 are considered to be periodic. Boundaries 3 and 4 are assigned to be no-flux for the 2D simulation. (b) We depict the upper part of the deforming lamella owing to the action of the shear flow pivoted around the point o , for a material line of initial width δ_0 and length \tilde{l} , deforming in the flow field so that its length increases to $\tilde{l}(t)$ and width reduces to $\delta(t)$. In the figure, the origin is fixed, the distance moved by the point p is the local velocity times the time. Considering a material line positioned at the center of the mixing zone in (a), we link the transport equations defined in the 2D system presented in (a) to a 1D transport process defined in the reference frame normal to the direction of deformation of the material line. The evolution of the system is then described as a function of a 1D coordinate x' which is normal to the stretching interface. The instantaneous angle with which the coordinate system has locally rotated can be found out through the arctan of the ratio of the lengths qr and or which are mentioned in the figure.

125 The transport of the reactant and product concentrations, \tilde{a} , \tilde{b} and \tilde{c} respectively (the symbols with tilde

126 represent dimensional quantities) is described by the following set of advection-reaction-diffusion equations

$$\begin{aligned}
 \frac{\partial \tilde{a}}{\partial \tilde{t}} + \tilde{\mathbf{u}} \cdot \nabla \tilde{a} &= D \nabla^2 \tilde{a} - k \tilde{a} \tilde{b}, \\
 \frac{\partial \tilde{b}}{\partial \tilde{t}} + \tilde{\mathbf{u}} \cdot \nabla \tilde{b} &= D \nabla^2 \tilde{b} - k \tilde{a} \tilde{b}, \\
 \frac{\partial \tilde{c}}{\partial \tilde{t}} + \tilde{\mathbf{u}} \cdot \nabla \tilde{c} &= D \nabla^2 \tilde{c} + k \tilde{a} \tilde{b},
 \end{aligned} \tag{1}$$

127 where D is a constant and represents the diffusion coefficient of the species.

128 Equation (1) is subjected to the following initial conditions:

$$\begin{aligned}
 \tilde{a}(\tilde{\mathbf{x}}, 0) &= \begin{cases} \tilde{n}_0 & \text{if } \tilde{x} < 0, \forall \tilde{y} \\ 0 & \text{otherwise} \end{cases}, \\
 \tilde{b}(\tilde{\mathbf{x}}, 0) &= \begin{cases} 0 & \text{if } \tilde{x} \geq 0, \forall \tilde{y} \\ \tilde{n}_0 & \text{otherwise} \end{cases}, \\
 \tilde{c}(\tilde{\mathbf{x}}, 0) &= 0 \quad \forall \tilde{\mathbf{x}}.
 \end{aligned} \tag{2}$$

129 The velocity field is given by $\tilde{\mathbf{u}} = G\tilde{y}\mathbf{e}_x$ with G representing the shear rate. The boundary conditions for
 130 the species A and B are that the top and bottom (walls 3 and 4) are no flux walls while the left and right
 131 walls (walls 1 and 2) are periodic (the effect of this periodicity is observed in the concentration distribution
 132 depicted in figure B.12 and figure B.12). These conditions are written down for solving for the species
 133 transport in a finite domain keeping in mind that we want to mimick a front where ideally the two domains
 134 are infinitely long. In such a system over a sufficient amount of time, there is no influence of time up until
 135 which the periodic image of the concentration interacts with itself. Beyond this time, the predictions of the
 136 2D simulations would no longer be valid for an isolated front but would represent the situation where there
 137 is overlapping of concentration profiles (boundary effects; this would imply longer times than those shown
 138 in figure B.12). There is, thus, no specific length scale attached to the problem; this is akin to most semi-
 139 infinite problems for diffusion, such as observed in heat transfer and mass transfer in a semi-infinite domain
 140 [85, 86]). Hence, to address the problem computationally for the 2D simulations, we have enforced periodic
 141 boundary conditions at the left and right boundaries in such a way that over the time of observation, the
 142 system does not experience the effects of the boundaries.

143 We nondimensionalize equation (1) by the following quantities:

$$\mathbf{x} = \tilde{\mathbf{x}}/\tilde{\delta}_0, t = \tilde{t}D/\tilde{\delta}_0^2, \mathbf{u} = \tilde{\mathbf{u}}/G\tilde{\delta}_0, \phi = \tilde{\phi}/\tilde{n}_0, \phi = a, b, c, \tag{3}$$

144 where in the system under consideration we choose the characteristic length to be of $\tilde{\delta}_0$, the characteristic
 145 time scale of the system based on the characteristic length, $\tilde{\delta}_0^2/D$, the characteristic velocity of the system
 146 is proportional to the rate of shear times the characteristic length. Proceeding based on this nondimension-

147 alization, and defining the Péclet and Damköhler numbers as,

$$\begin{aligned} \text{Pe} &= \frac{\tau_D}{\tau_a} = \frac{G\tilde{\delta}_0^2}{D}, \\ \text{Da} &= \frac{\tau_D}{\tau_r} = \frac{k\tilde{n}_0\tilde{\delta}_0^2}{D}, \end{aligned} \quad (4)$$

148 which represent respectively the ratio of the typical diffusion time scale $\tau_D = \tilde{\delta}_0^2/D$ to the typical advection
 149 time scale $\tau_a = G^{-1}$ and that of the typical diffusion time scale to the typical reaction time scale respectively
 150 $\tau_r = (k\tilde{n}_0)^{-1}$, equations (1) and (2) may be then recast as:

$$\begin{aligned} \frac{\partial a}{\partial t} + \text{Pe} \mathbf{u} \cdot \nabla a &= \nabla^2 a - \text{Da} a b, \\ \frac{\partial b}{\partial t} + \text{Pe} \mathbf{u} \cdot \nabla b &= \nabla^2 b - \text{Da} a b, \\ \frac{\partial c}{\partial t} + \text{Pe} \mathbf{u} \cdot \nabla c &= \nabla^2 c + \text{Da} a b, \end{aligned} \quad (5)$$

151 subjected to the initial conditions

$$\begin{aligned} a(\mathbf{x}, 0) &= \begin{cases} 1 & \text{if } x < 0, \forall y \\ 0 & \text{otherwise} \end{cases}, \\ b(\mathbf{x}, 0) &= \begin{cases} 0 & \text{if } x \geq 0, \forall y \\ 1 & \text{otherwise} \end{cases}, \\ c(\mathbf{x}, 0) &= 0 \quad \forall \mathbf{x}. \end{aligned} \quad (6)$$

152 As we shall see later, depending on the relative strengths of Pe and Da the system evolution exhibits markedly
 153 different dynamics.

154 Note that the scenario where there is no definite length scale associated with the particular problem
 155 under consideration can be tackled in the following manner. A system lacking a characteristic lengthscale
 156 is akin to analysis pertaining to semi-infinite domains where the pertinent lengthscale must be obtained
 157 by a combination of the physically relevant parameters in the system [87]. In the context of the present
 158 problem, we may define two length scales based on the combination of the diffusion coefficient, D , the
 159 reaction kinetics constant, k , and the average shear rate, G . If the system is thought to evolve based on the
 160 balance of diffusion and reaction kinetics the length scale $\tilde{\delta}_0$ may be recast as $\sqrt{D/(kn_0)}$, which is obtained
 161 by comparing the characteristic lengthscale of molecular diffusion during the time τ_r , i.e. $\sqrt{D\tau_r}$, over the
 162 the characteristic reaction time $\tau_r = 1/(kn_0)$. On the other hand, if the system evolves based on the balance
 163 of diffusion and stretching, the lengthscale $\tilde{\delta}_0$ may be recast as $\sqrt{D/G}$, which is obtained by comparing the
 164 characteristic diffusion lengthscale to the stretching time $\tau_s = 1/G$. In the present derivation, we do not
 165 restrict ourselves to the choice of $\tilde{\delta}_0$ thereby allowing us to generalize the analysis presented here.

166 *2.2. Reaction-diffusion system in a Lagrangian frame*

167 To derive analytical expressions quantifying the coupling between reaction, diffusion and shear , we
 168 simplify the equations and make them more tractable by reducing the dimensionality of the problem [56, 88,
 169 89, 54]. In order to provide a better insight into the fundamental process of stretching augmented reactive
 170 mixing, we convert the 2D transport problem defined in the laboratory frame to a one-dimensional (1D)
 171 transport problem defined in the local Lagrangian frame attached to a particular material point and rotating
 172 with the flow. A material line is a purely kinematic quantity, independent of the species residing in it [56],
 173 but we choose the material line that coincides with the middle line of the mixing zone (see Figure 1), which
 174 is initially oriented along the direction y . Considering a volume of initial constant thickness $\tilde{\delta}_0$ around the
 175 material line (also referred to as a lamella, see figure 1), the elongation of the line by shear deformation $\tilde{\rho}(t)$
 176 leads to a simultaneous compression of the thickness $\tilde{\delta}$ (owing to incompressibility), such as [56, 90]. We
 177 therefore confine our attention to the region at the interface of the two reactants and track it as it evolves
 178 with the flow.

179 The advection-diffusion-reaction equation (1) for species \tilde{a} may be simplified by expressing it in the
 180 coordinate system (\tilde{x}', \tilde{y}') attached to a material line (Figure 1) [e.g., 56] . The simultaneous effect of
 181 compression, diffusion and reaction is quantified in a Lagrangian frame as follows (see Appendix A for the
 182 detailed derivation):

$$\frac{\partial \tilde{a}}{\partial \tilde{t}} - \Omega \tilde{x}' \frac{\partial \tilde{a}}{\partial \tilde{x}'} = D \frac{\partial^2 \tilde{a}}{\partial \tilde{x}'^2} - k \tilde{a} \tilde{b}, \quad (7)$$

and correspondingly for species \tilde{b} and \tilde{c} . We have defined here the stretching rate

$$\Omega = -\frac{1}{\tilde{\delta}} \frac{d\tilde{\delta}}{d\tilde{t}}, \quad (8)$$

183 where $\tilde{\delta}$ is the width of a material strip whose initial orientation is perpendicular to the flow direction and
 184 whose initial length is $\tilde{\delta}_0$ (see Appendix A),

$$\tilde{\delta} = \frac{\tilde{\delta}_0}{\sqrt{1 + G^2 \tilde{t}^2}}. \quad (9)$$

185 It is observed in equation (7) that species evolution is only in the direction transverse to the elongation,
 186 that is \tilde{x}' . This stems from the fact that for a linear shear, concentration gradients are zero in the direction
 187 of elongation and exist only in the direction of compression [19, 56]. Even for arbitrary flows equation (7)
 188 is still a good approximation since concentration gradients are continuously maximized in the direction of
 189 compression [54, 66] (Please refer to Appendix A for a discussion).

190 We can make further progress by transforming the above equation into a diffusion-reaction system by
 191 making use of the following rescaled variables [56]:

$$z = \frac{\tilde{x}'}{\tilde{\delta}} \quad \text{and} \quad \theta = \int_0^{\tilde{t}} d\tau \frac{D}{\tilde{\delta}(\tau)^2}. \quad (10)$$

192 where θ is referred to as the warped time which represents the integral diffusion time over the lamella
 193 thickness. The transformed spatial coordinate, z , must not be confused with the z coordinate in the usual
 194 orthogonal three-dimensional Cartesian system. We obtain:

$$\begin{aligned}\frac{\partial \tilde{a}}{\partial \theta} &= \frac{\partial^2 \tilde{a}}{\partial z^2} - k\tilde{a}\tilde{b}\frac{\tilde{\delta}^2}{D}, \\ \frac{\partial \tilde{b}}{\partial \theta} &= \frac{\partial^2 \tilde{b}}{\partial z^2} - k\tilde{a}\tilde{b}\frac{\tilde{\delta}^2}{D}, \\ \frac{\partial \tilde{c}}{\partial \theta} &= \frac{\partial^2 \tilde{c}}{\partial z^2} + k\tilde{a}\tilde{b}\frac{\tilde{\delta}^2}{D}.\end{aligned}\tag{11}$$

195 Furthermore, a nondimensionalization may be employed for the lamella thickness as $\delta = \tilde{\delta}/\tilde{\delta}_0$, and by taking
 196 advantage of equation (8) we may simplify the definition of the warped time to obtain the form

$$\theta = \int_0^{\tilde{t}} \frac{d\tau}{\tilde{\delta}(\tau)^2/D} = \frac{D}{\tilde{\delta}_0^2} \int_0^{\tilde{t}} d\tau (1 + G^2\tau^2) = \frac{D}{\tilde{\delta}_0^2} \left(\tilde{t} + \frac{G^2\tilde{t}^3}{3} \right) .\tag{12}$$

197 where we have used the fact that at $\tilde{t} = 0$, $\theta = 0$. Using this, and the aforementioned nondimensional
 198 scheme, we may write the governing equations in the Lagrangian frame for the three species in the frame
 199 attached with the deforming material line as

$$\begin{aligned}\frac{\partial a}{\partial \theta} &= \frac{\partial^2 a}{\partial z^2} - \text{Da} ab\delta^2, \\ \frac{\partial b}{\partial \theta} &= \frac{\partial^2 b}{\partial z^2} - \text{Da} ab\delta^2, \\ \frac{\partial c}{\partial \theta} &= \frac{\partial^2 c}{\partial z^2} + \text{Da} ab\delta^2,\end{aligned}\tag{13}$$

200 where the Péclet number and Damköhler numbers have already been defined in section 2.1 and

$$\delta = \frac{1}{\sqrt{1 + \text{Pe}^2 t^2}}\tag{14}$$

201 from equation (9).

202 The solution for the set of equations (11) yields the temporal and spatial evolution for the concentration
 203 fields of the different species. The initial conditions specified for solving equation (11) are that as $z \rightarrow -\infty$,
 204 $a = 1, b = 0$ and that as $z \rightarrow \infty$, $b = 1, a = 0$. The aim of the present work is to depict how the chemical
 205 kinetics constants in conjunction with the lamella stretching impact the mass of the product formed in such
 206 a scenario.

207 We may write the mass of the product as $\tilde{m}_c = \int_{-\infty}^{\infty} d\tilde{x}' \tilde{c}(x')l(t)$, where $l(t)$ represents the length of the
 208 interface. In a nondimensional sense, it may be recast as

$$m_c = \frac{\tilde{m}_c}{n_0 l_0 \tilde{\delta}_0} = \int_{-\infty}^{\infty} dz c(z)l\delta = \int_{-\infty}^{\infty} dz c(z) ,\tag{15}$$

209 where we have made use of the fact that $l\delta = 1$ (owing to the incompressibility condition which imposes
 210 that the amount of longitudinal stretching be the inverse of the amount of transverse compression in order
 211 to conserve the material volume).

212 Another quantity of interest is the reactive mixing scale, which quantifies the spatial localization of
 213 chemical reactivity in a mixing front [27]. A small reactive mixing scale denotes the existence of localized
 214 hotspots of chemical reaction, while a large reactive mixing scale implies that the reaction zone is diffuse.
 215 The reactive mixing scale is thus an important characteristic of reactive fronts, which, as discussed in the
 216 following, depends non-trivially on Pe , Da and on the observation time. The reactive mixing scale can be
 217 estimated from the width of the reaction front from the second moment of the reaction rate across the front
 218 as

$$\tilde{s}_r = \left(\frac{\int_{-\infty}^{\infty} d\tilde{x}' \tilde{x}'^2 Da ab}{\int_{-\infty}^{\infty} d\tilde{x}' Da ab} \right)^{1/2}, \quad (16)$$

219 In a nondimensional sense, it can be written in terms of the integral in the transformed coordinate, z , as

$$s_r = \left(\frac{\int_{-\infty}^{\infty} dz z^2 Da ab}{\int_{-\infty}^{\infty} dz Da ab} \right)^{1/2} \delta. \quad (17)$$

220 Note that the reactive mixing scale is different from the conservative mixing scale [91, 66] since it is affected by
 221 chemical reactions that tend to reduce it through consumption of reactants. Besides the temporal evolution
 222 of the mass of the product, the temporal evolution of the width of the reaction zone is also of interest and
 223 is analyzed in section 4. The initial reaction width, i.e. $s_r(0)$, will be referred to as s_0 .

224 3. Analytical predictions for the temporal evolution of the mass of product

225 Having discussed the general 1D framework used to investigate the transport of reactive species, we focus
 226 on the various limits of the temporal evolution of the mass of product. Let us remind here that the Péclet
 227 number denotes the ratio of the typical diffusion time scale (over the distance δ_0) to the typical advection
 228 time scale (due to the fluid shear), while the Damköhler number denotes the ratio of the typical reaction
 229 time scale to the typical diffusion time scale. This implies that the dimensionless time $t = \tilde{t}/\tau_D$ allows us to
 230 distinguish the dynamics of the systems in terms of the relative values of $Pe^{-1} = \tau_a/\tau_D$ and $Da^{-1} = \tau_r/\tau_D$.

231 The first scenario, termed as *weak stretching* scenario, is defined by a system for which $Da^{-1} < Pe^{-1}$. In
 232 such a scenario, 3 different time regimes can be distinguished. The first regime occurs for a dimensionless time
 233 $t \ll Da^{-1}$. Here the reaction dynamics is determined by the interaction of diffusion and reaction kinetics
 234 since the time is small compared to the characteristic reaction time. In the second regime, occurring for
 235 $Da^{-1} < t < Pe^{-1}$, the reaction is diffusion limited since the time is large compared to the reaction time (i.e.
 236 reactions are fast) but small compared to the characteristic shear time (i.e. stretching does not play a role
 237 yet). In the asymptotic long time regime, $t \gg Pe^{-1}$, the shear action of the flow field is activated and the
 238 reaction kinetics is controlled by stretching-enhanced diffusion.

239 The second scenario, termed as the *strong stretching* scenario is defined as $Pe^{-1} < Da^{-1}$. The first
 240 regime, characterized by a dimensionless time $t \ll Pe^{-1}$ is similar to the first regime in the weak stretching

241 case. The system is dominated by the interaction of diffusion and reaction kinetics. The second time regime,
 242 for which $\text{Pe}^{-1} < \text{Da}^{-1}$, sees a reaction behavior that results from the interaction of the reaction kinetics
 243 and stretching enhanced diffusion as time is large enough for stretching to be activated but small compared
 244 to the characteristic reaction time. This regime is particularly interesting as stretching and reaction kinetics
 245 are fully coupled. In the long time regime, $t \gg \text{Da}^{-1}$, the reaction behavior is fully limited by stretching-
 246 enhanced diffusion as reaction kinetics is no longer limiting.

247 As detailed in the introduction, reaction front kinetics have been investigated mostly in the limits $t \ll$
 248 Pe^{-1} (no shear) or $t \gg \text{Da}^{-1}$ (fast reactions under shear), which represents only a subset of the range of
 249 possible regimes described above. In the following we derive the temporal evolution of the product mass for
 250 all the aforementioned regimes.

251 3.1. Weak stretching scenario: $\text{Da}^{-1} < \text{Pe}^{-1}$

252 For a dimensionless time $t \ll \text{Pe}^{-1}$ shear does not affect the reactive transport behavior as both the
 253 elongation ρ and the width δ are approximately constant (see equation (9)). Thus, the evolution of the
 254 product c can be represented by the diffusion reaction equation

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial x^2} + \text{Da}ab. \quad (18)$$

255 Hence, this regime is akin to the case where there is no imposed flow field [27]. Conversely for $t \gg \text{Pe}^{-1}$, shear
 256 plays an important role in increasing the area available for diffusive mass transfer, and enhancing chemical
 257 gradients by compression. In the following, we discuss the three regimes separated by the characteristic
 258 times Da^{-1} and Pe^{-1} .

259 3.1.1. Reaction-diffusion regime (negligible shear): $t \ll \text{Da}^{-1}$

260 In this regime, the initial profiles of a and b both evolve due to diffusion while the impact of reaction
 261 is still weak. In such a case, the concentration profiles of the two reactants can be approximated by the
 262 diffusive profiles [85]

$$a = \frac{1}{2}(1 + \text{erf}(x/2\sqrt{t})), \quad (19)$$

$$b = \frac{1}{2}(1 - \text{erf}(x/2\sqrt{t})), \quad (20)$$

263 Thus, in this regime where we can neglect the effect of shear, we may write the evolution of c , inserting
 264 equation (19) and (20) in equation (18), as

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial x^2} + \text{Da} \frac{1}{4} \left[1 - \text{erf} \left(\frac{x}{2\sqrt{t}} \right)^2 \right] \quad (21)$$

265 which, in the vicinity of the interface, i.e. the origin (where most of the product is formed) may be further
 266 simplified as

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial x^2} + \text{Da} \frac{1}{4}. \quad (22)$$

267 We may integrate this over space to obtain the temporal evolution of the mass of product formed as

$$\frac{\partial m_c}{\partial t} = \left[\frac{\partial c}{\partial x} \right]_{-\infty}^{\infty} + \frac{\text{Da}}{4} s_r \quad (23)$$

268 wherein the second term on the right hand side has a contribution from $\text{Da}/4$ which is concentrated near
 269 the interface (at the origin), while s_r represents the reactive mixing scale. The first term on the right hand
 270 size is zero on account of the fact that the concentration profile decays away from the front towards either
 271 reactant so that $\partial c/\partial x(x \rightarrow \pm\infty) = 0$. Now, at such times where $t \ll \text{Pe}^{-1}$ and $t \ll \text{Da}^{-1}$, the growth of
 272 the reactive mixing scale is only *diffusion controlled*, which leads to the growth of the reactive mixing scale
 273 as $s_r \sim \sqrt{t}$. Thus, we arrive at the evolution of m_c by integrating equation (23) in time as

$$m_c \approx \frac{\text{Da}}{4} t^{3/2}. \quad (24)$$

274 This result is equivalent to that obtained by Arshadi and Rajaram [29] in the early time regime of a
 275 reaction front with no shear.

276 3.1.2. Diffusion limited regime (negligible shear at intermediate times): $\text{Da}^{-1} < t < \text{Pe}^{-1}$

277 In order to determine the behavior at longer times (where the reactions are fast and the influence of
 278 the shear is still not present; this regime is similar to the case at long times in the absence of any imposed
 279 flow), we follow the lead of Larralde et al. [27]. For completeness, we summarize here the main steps of the
 280 derivation and the key results. We begin by representing the concentration of the product as a perturbation
 281 on the diffusive case. Formally, we first observe that the concentrations of the two reactants may be written
 282 as $a = F + g$ and $b = g$ where $F = a - b$ satisfies the conservative equation (by noting that subtracting the
 283 two concentration fields eliminates the nonlinearity of the reaction term)

$$\frac{\partial F}{\partial t} = \frac{\partial^2 F}{\partial x^2} \quad (25)$$

284 where g represents the concentration perturbation. Consequently, the governing equation for g may be
 285 represented as

$$\frac{\partial g}{\partial t} = \frac{\partial^2 g}{\partial x^2} - \text{Da} g \left(\text{erf} \left(\frac{x}{\sqrt{4t}} \right) + g \right) \quad (26)$$

286 which may be simplified by neglecting the nonlinear contribution from g^2 (since g is considered to be
 287 a perturbation to F) and linearizing $\text{erf}(x/\sqrt{4t})$ for times so large and the point of interest such that
 288 $x/\sqrt{4t} \ll 1$, we obtain

$$\frac{\partial g}{\partial t} \approx \frac{\partial^2 g}{\partial x^2} - \text{Da} g \frac{x}{\sqrt{\pi t}} \quad (27)$$

289 At times sufficiently long for the temporal derivative to be negligible, the time behaves as a parameter to
 290 the Airy differential equation given by: $\frac{\partial^2 g}{\partial x^2} - \text{Da} g \frac{x}{\sqrt{\pi t}} = 0$. The solution to this may be written by resorting
 291 to an Airy function, as [27]:

$$g \sim f(t) \text{Ai} \left(\lambda \frac{x}{t^{1/6}} \right), \quad \lambda = \left(\frac{\text{Da}}{\sqrt{\pi}} \right)^{1/3} \quad (28)$$

292 We may assume a power-law representation for the function $f(t)$ so that the form for the concentration
 293 perturbation is represented as $g = \psi t^\alpha Ai(\lambda x/t^{1/6})$. By equating the form of the nonlinear term in (26) and
 294 the spatial derivative term on the right hand side of equation (28), we see that $\alpha = -1/3$ and $\psi = 1/\lambda$.
 295 Thus, the perturbation to the concentration is found out as

$$g \sim \frac{t^{-1/3}}{\lambda} Ai\left(\frac{\lambda x}{t^{1/6}}\right) . \quad (29)$$

296 The above may be further simplified for the case of large enough x such that $\lambda x \gg t^{1/6}$ to yield

$$g \sim \frac{t^{-1/3}}{\lambda} \left(\frac{\lambda x}{t^{1/6}}\right)^{-1/4} \exp\left(-\frac{2}{3} \left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right) , \quad (30)$$

297 while the reaction term may be recast based on Eq.(27) to yield

$$R \sim \text{Da} \frac{x}{\sqrt{t}} \frac{t^{-1/3}}{\lambda} \left(\frac{\lambda x}{t^{1/6}}\right)^{-1/4} \exp\left(-\frac{2}{3} \left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right) . \quad (31)$$

298 The time integral of the reaction term may then be obtained as $I \sim \int_0^\infty R dt$, which, according to equa-
 299 tion (31) can be written as

$$I \sim \int_0^\infty dt \text{Da} \frac{x}{\sqrt{t}} \frac{t^{-1/3}}{\lambda} \left(\frac{\lambda x}{t^{1/6}}\right)^{-1/4} \exp\left(-\frac{2}{3} \left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right) . \quad (32)$$

300 Upon simplification, equation (32) can be written, in leading order, as

$$I \sim \lambda t^{1/3} \left(\frac{\lambda x}{t^{1/6}}\right)^{-3/4} \exp\left(-\frac{2}{3} \left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right) \quad (33)$$

Towards determining the mass of product, we appeal to the above equation and equation (15) to obtain

$$m_c = 2 \int_0^\infty dx \lambda t^{1/3} \left(\frac{\lambda x}{t^{1/6}}\right)^{-3/4} \exp\left(-\frac{2}{3} \left(\frac{\lambda x}{t^{1/6}}\right)^{3/2}\right)$$

301 where the factor of 2 appears when the spatial integration is done from the lower bound 0 instead of $-\infty$
 302 while keeping in mind that the integral is symmetric for the concentration perturbation. The integral can
 303 be evaluated by making the change of variable $\beta = \lambda x/t^{1/6}$ to obtain

$$m_c \approx t^{1/2} 2 \int_0^\infty d\beta \exp\left(\frac{2}{3} \beta^{3/2}\right) \beta^{-3/4} \approx 8 t^{1/2} . \quad (34)$$

304 The general observation is that the temporal evolution of the mass of the product formed in this regime
 305 becomes independent of the Damköhler number, which is expected for this regime that is purely diffusion-
 306 limited. We note here that the integral appearing in equation (34) comes out to be $2 \int_0^\infty dx \exp(2x^{3/2}/3)x^{-3/4} =$
 307 $2/3 \times (2^{5/6} 3^{1/6} \pi) \Gamma(5/6)$ whose approximate value is 7.94, and which we further approximate as 8 in the
 308 equation above. This result is in line with the derivation of Larralde [27] and of Arshadi and Rajaram [29]
 309 for a reactive front with no shear.

310 *3.1.3. Shear-enhanced reactive mixing regime: $t \gg \text{Pe}^{-1}$*

311 We now turn our attention to the case where the imposed velocity gradient has significant bearing on
 312 the dynamics of the reaction-diffusion system. Towards analyzing this, we begin with equation (11) and
 313 move ahead with the logic similar to that employed in section 3.1.2. In $z - \theta$ coordinates, which account for
 314 the effect of shear-induced elongation and compression of the interface, we may write the evolution of the
 315 perturbation to the concentrations of a and b , as in equation (26), as

$$\frac{\partial g}{\partial \theta} = \frac{\partial^2 g}{\partial z^2} - \frac{\text{Da}}{1 + \text{Pe}^2 t^2} g \left(\text{erf} \left(\frac{z}{\sqrt{4\theta}} \right) + g \right), \quad (35)$$

316 which can be further simplified by neglecting the nonlinear contribution of the g^2 term to yield

$$\frac{\partial g}{\partial \theta} = \frac{\partial^2 g}{\partial z^2} - \frac{\text{Da}}{1 + \text{Pe}^2 t^2} g \frac{z}{\sqrt{\pi\theta}}. \quad (36)$$

317 For $t > \text{Pe}^{-1}$, we can approximate $1/1 + \text{Pe}^2 t^2 \approx 1/\text{Pe}^2 t^2$. We consider the system in the x' and t coordinates
 318 which are easily transformed by recalling that $z = x'/\delta \sim x' \text{Pe} t$ and $\theta \sim \text{Pe}^2 t^3/3$, obtaining

$$\frac{\partial g}{\partial t} = \frac{\partial^2 g}{\partial x'^2} - \text{Da} \sqrt{\frac{3}{\pi}} g \frac{x'}{t}. \quad (37)$$

319 The solution to equation (37) may be obtained (in analogous manner to that in section 3.1.2) as

$$g \approx \frac{t^{-1/3}}{\lambda'} \text{Ai} \left(\frac{\lambda' x'}{t^{1/6}} \right), \quad (38)$$

320 where $\lambda' = (\text{Da} \sqrt{3/\pi})^{1/3}$. Proceeding, we may write the rate of reaction term as

$$R = \text{Da} \frac{x'}{t^{1/2}} g \approx \lambda'^3 \frac{x'}{t^{1/2}} \times \frac{t^{-1/3}}{\lambda'} \text{Ai} \left(\frac{\lambda' x'}{t^{1/6}} \right), \quad (39)$$

321 which we may simplify to obtain

$$R \approx \lambda' \left(\frac{\lambda' x'}{t^{1/6}} \right) t^{-2/3} \text{Ai} \left(\frac{\lambda' x'}{t^{1/6}} \right). \quad (40)$$

322 Proceeding further, we may integrate the above expression in space and time to obtain

$$m_c \approx \lambda' t^{1/3} 2 \int_0^\infty dz \left(\frac{\lambda' x'}{t^{1/6}} \right)^{-3/4} \exp \left(-\frac{2}{3} \left(\frac{\lambda' x'}{t^{1/6}} \right)^{3/2} \right), \quad (41)$$

323 the evaluation of the integral over the z space necessitates converting the groups of the variables into the
 324 form of $\gamma = (\lambda z)/(\text{Pe} t^{7/6})$ (the conversion of the variable from x' in terms of z results in the appearance of
 325 Pe through the relation $z \sim x' \text{Pe} t$ as noted earlier). Performing this, we obtain from equation (41) that

$$m_c \approx 8 \text{Pe} t^{3/2}. \quad (42)$$

326 This expression shows that, as expected, the impact of the shear action of the flow leads to a stronger
 327 increase in the product mass than that due to pure diffusion as given by equation (34). This result is
 328 validated against numerical simulations in section 6.

329 It is interesting to note however that the scaling is the same as in the kinetics limited regime (equation
330 25). For the weak stretching scenario, discussed in this section, the effects of kinetics limitation, due to finite
331 reaction times, are confined to the domain of dimensionless times $t \ll \text{Da}^{-1}$, while the effects of mixing
332 enhancement by shear are confined to the domain $t \gg \text{Pe}^{-1}$. Hence, as $\text{Da}^{-1} < \text{Pe}^{-1}$, these two domains
333 are disjoint and these two effects are decoupled. As discussed in the next section, this is not the case for the
334 strong stretching scenario, which leads to new scaling laws.

335 3.2. Strong stretching scenario: $\text{Pe}^{-1} < \text{Da}^{-1}$

336 In the strong stretching scenario, the shear time scale Pe^{-1} is smaller than the characteristic reaction
337 scale Da^{-1} . In this scenario, the diffusion-limited reaction regime $\text{Da}^{-1} < t < \text{Pe}^{-1}$ does not exist. Instead,
338 we find a new regime characterized by the interaction of stretching-enhanced mixing and reaction. It must
339 first be noted that for a dimensionless time $t \ll \text{Pe}^{-1}$ the effect of stretching, as manifested through the
340 presence of the term δ^2 in the reaction term (where we recall that $\delta = 1/\sqrt{1 + \text{Pe}^2 t^2}$), is weak, and thus
341 the approximate equations assuming $1 + \text{Pe}^2 t^2 \approx 1$ result in the analysis seen in section 3.1.1. Essentially
342 this implies that in the initial time, the system behaves as if there is no imposed shear. On the other hand,
343 in the long time regime ($t \gg \text{Da}^{-1}$), the reaction behavior is the same as the one obtained in section 3.1.3,
344 i.e. that of shear-enhanced reactive mixing with no kinetics. In the following, we therefore only study the
345 reaction behavior in the intermediate regime ($\text{Pe}^{-1} < t < \text{Da}^{-1}$) in which the effect of flow stretching and
346 kinetics limitations are both non-negligible. We thus begin the analysis through equation (13) in terms of
347 $z - \theta$ coordinates:

$$\frac{\partial c}{\partial \theta} = \frac{\partial^2 c}{\partial z^2} + \text{Da} a b \delta^2 \quad , \quad (43)$$

348 As in section 3.1.1, approximating the profiles of a and b by diffusive profiles, which are weakly affected by
349 reaction, and linearizing close to $z = 0$, this expression can be integrated over z and simplified to obtain
350 (using the arguments that $\partial c / \partial z(z \rightarrow \pm\infty) = 0$)

$$\frac{\partial m_c}{\partial \theta} \approx \frac{\text{Da}}{4} \delta^2 s_r \quad . \quad (44)$$

351 Note that this expression is based on the same approximations as equation (23) but it is here written in
352 $\{\theta, z\}$ coordinates to account for shear. Recalling that $d\theta/dt = 1/\delta^2$, we can rewrite equation (44) as

$$\frac{\partial m_c}{\partial \theta} \approx \frac{\text{Da}}{4} \left(\frac{d\theta}{dt} \right)^{-1} s_r \implies \frac{\partial m_c}{\partial t} \approx \frac{\text{Da}}{4} \sqrt{\theta} \quad . \quad (45)$$

353 where we have made use of the fact that during this warped time, the diffusive growth of the width is
354 proportional to $\sqrt{\theta}$. By utilizing the form of the warped time, $\theta = (t + \text{Pe}^2 t^3/3) \sim \text{Pe}^2 t^3/3$, we finally
355 obtain

$$\frac{\partial m_c}{\partial t} \approx \frac{\text{DaPe}}{4\sqrt{3}} t^{3/2} \implies m_c \approx \frac{\text{DaPe}}{10\sqrt{3}} t^{5/2} \quad . \quad (46)$$

356 Hence, it is observed that the coupling of shear-enhanced mixing and kinetics limitations leads to a strong
 357 acceleration of the effective kinetics, which is faster than all previously known regimes. Consistently, the
 358 mass produced is proportional to both Da and Pe .

359 Figure 2 synthesizes the different regimes expected from the analysis for the scaling of the mass of
 360 produced in mixing fronts. The new coupled stretching and kinetics regime, that shows accelerated mass
 361 production $m_c \sim PeDa t^{5/2}$, appears to cover a significant part of the diagram. The presented analytical
 362 derivations thus provide a unified theoretical framework covering the full space of Pe , Da and t parameters.

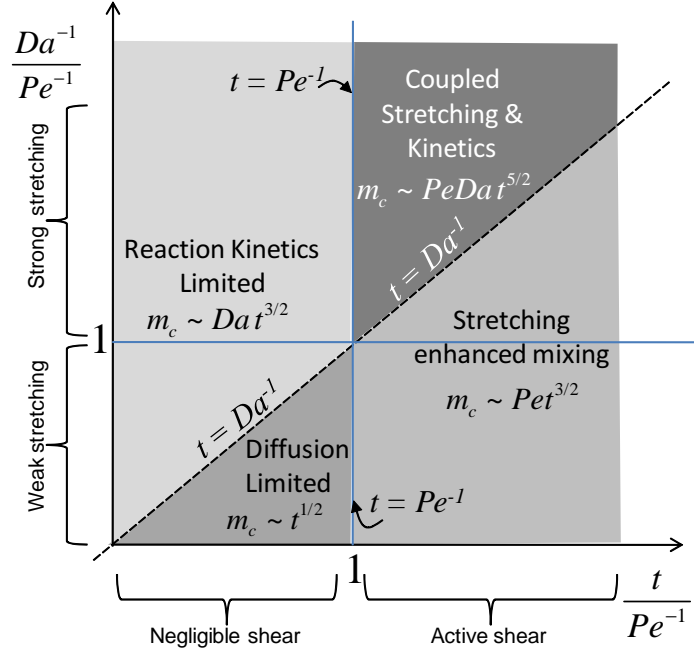


Figure 2: Diagram synthesizing the different regimes predicted for the scaling of the mass of product. The y -axis represents the two vertical separations of weak stretching and strong stretching when $Da^{-1}/Pe^{-1} < 1$ and > 1 respectively. The x -axis is also demarcated by the regimes of negligible shear and strong shear through $t/Pe^{-1} < 1$ and > 1 respectively. We recall that t is the dimensionless time $t = \tilde{t}D/\delta_0^2$.

363 3.3. Transition times

364 In this section, we quantify more precisely the transition times between the different regimes discussed
 365 above. Quite naturally, in the absence of any imposed shear flow, we would expect the presence of only one
 366 dimensionless transition time at Da^{-1} between the kinetics-limited regime, discussed in section 3.1.1, and
 367 the diffusion-limited regime, discussed in section 3.1.2. The interest of the problem under consideration lies
 368 in configurations with significant imposed stretching. Depending on the relative strength of the stretching
 369 and reaction kinetics, we may have various transitions occurring as time progresses. We shall discuss these
 370 in details below.

371 Let us first consider the scenario of weak stretching $\text{Da}^{-1} < \text{Pe}^{-1}$. In this case, the system is expected
372 to have two transitions. The first transition occurs between the early time kinetics-limited regime and the
373 intermediate diffusion-limited regime, while the second transition occurs between the intermediate diffusion-
374 limited reaction regime and the long time shear-enhanced reactive mixing regime. Towards determining
375 the first transition time we must have an overlap of the product formed between these regions so that
376 $\text{Da}/4t^{3/2} = 8t^{1/2}$ (see sections 3.1.1 and 3.1.2)). This yields the dimensionless transition time as $t_{\text{Da}_1} =$
377 $32/\text{Da}$. Consequently, the mass of product at this transition may be found out as $m_{c,\text{Da}_1} = 45/\sqrt{\text{Da}}$.
378 Moving on to the second transition from the diffusion-limited reaction regime (section 3.1.2) to the long
379 time shear-mixing-limited reaction regime (section 3.1.3), we equate the masses of product for the two
380 regimes, obtaining $8t^{1/2} = 8\text{Pe}t^{3/2}$, which yields a characteristic dimensionless time $t_{\text{Pe}_1} = \text{Pe}^{-1}$. In this
381 case the mass of product at the transition is given by $m_{c,\text{Pe}_1} = 8\text{Pe}^{-1/2}$.

382 In the second scenario, we consider the situation with strong imposed stretching $\text{Da}^{-1} > \text{Pe}^{-1}$. In
383 this scenario, we still expect to have two transitions. The first transition will be from the kinetics-limited
384 regime to the intermediate coupled stretching enhanced mixing and kinetics limited regime while the second
385 transition will be from this intermediate regime to the long time shear-enhanced mixing regime with no
386 kinetics limitation. At the first transition time, we must have an equality of the masses of product determined
387 in the two regimes $\frac{\text{Da}}{4}t^{3/2} = \frac{\text{PeDa}}{10\sqrt{3}}t^{5/2}$, which yields a dimensionless transition time $t_{\text{Pe}_2} = \frac{1}{\text{Pe}}\frac{5\sqrt{3}}{2}$; at this time
388 the mass of the product is found out to be $m_{c,\text{Pe}_2} = \frac{\text{Da}}{\text{Pe}^{3/2}}\frac{1}{4}\left(\frac{5\sqrt{3}}{2}\right)^{3/2}$. Similarly, at the second transition,
389 we have $\text{PeDa}\frac{t^{5/2}}{10\sqrt{3}} = 8\text{Pe}t^{3/2}$. The resulting characteristic transition time is $t_{\text{Da}_2} = \frac{1}{\text{Da}}80\sqrt{3}$, corresponding
390 to a mass of product at the dimensionless transition given by $m_{c,\text{Da}_2} = 8\frac{\text{Pe}}{\text{Da}^{3/2}}(80\sqrt{3})^{3/2}$.

391 4. Temporal dynamics of the reactive mixing scale

392 We now consider the temporal behavior of the reactive mixing scale s_r , which characterizes the spatial
393 extent of the zone where reactions take place (equation (16)). The mixing of two separated reactants creates
394 a hotspot of reaction at the interface, which may be highly localized in space. This can be due to i) slow
395 diffusion, ii) fast reactions, which implies that reactants are immediately consumed as they interpenetrate
396 each other, and iii) compression of the interface due to shear deformation. In the following, we quantify the
397 interplay of these different processes that determines the evolution of the reactive mixing scale.

398 4.1. Dynamics of the reactive mixing scale in the absence of shear flow

399 For the scenario where there is no imposed velocity gradient, the evolution of the reactive mixing scale
400 at sufficiently long times has been mathematically demonstrated earlier [27]. For completeness, we mention
401 it here. It was seen in 3.1.1 that the initial scaling of the reactive mixing scale, for $t < \text{Da}^{-1}$, is expected
402 to be diffusion controlled since reactions are too slow to affect the diffusive growth of the interpenetration

403 zone of the two reactants, resulting in

$$s_r \sim \sqrt{t}. \quad (47)$$

404 After this first regime, for $t > \text{Da}^{-1}$, reactions can be considered to be fast compared to the observation time.
 405 Hence, the progression of the reactive mixing scale is impeded by consumption of the diffusive reactants.
 406 Therefore it evolves at a much slower rate. The temporal scaling of the reactive mixing scale can be estimated
 407 by considering the form of the reaction rate in equation (40), which follows a scaling form $R = h(t)f(x/t^{1/6})$.
 408 In this case, the similarity variable is $x/t^{1/6}$ and the reactive mixing scale, defined by the second moment
 409 of R in equation (16), is expected to follow a scaling [27]

$$s_r \sim t^{1/6}. \quad (48)$$

410 4.2. Reactive mixing scale dynamics with imposed flow: strong stretching regime, $\text{Pe}^{-1} < \text{Da}^{-1}$

411 In the scenario of strong stretching, we focus on times larger than Pe^{-1} because before this characteristic
 412 time the reactive mixing scale is expected to be diffusion controlled, a case which has been discussed above.
 413 For times $t < \text{Da}^{-1}$, reaction can be considered to be too slow to affect the progression of the reactive mixing
 414 scale. Hence, we can apply the same approximation as in the above case but in the $z - \theta$ coordinates, which
 415 account for shear deformation. In analogy to equation (47), the reactive mixing scale is expected to grow as
 416 $z_r \sim \sqrt{\theta}$ (diffusion-limited growth) where z_r denotes the reactive mixing scale in the $z - \theta$ coordinates (we
 417 recall that $z = x'/\delta$). Therefore, the reactive mixing scale in the x' coordinate is

$$s_r \sim \delta\sqrt{\theta}. \quad (49)$$

418 We utilize the above functional relationship to arrive at a governing equation for the width. Therefore, we
 419 differentiate this with respect to time, t , to obtain

$$\frac{1}{s_r} \frac{\partial s_r}{\partial t} = \frac{1}{\delta} \frac{\partial \delta}{\partial t} + \frac{1}{2s_r^2}. \quad (50)$$

420 The above equation governs the compression and diffusion of a front for the case of strong stretching at
 421 times $t < \text{Da}^{-1}$. For $t > \text{Da}^{-1}$, the expected scaling is the same as discussed in the previous section at long
 422 times (equation (48)).

423 Note that equation (50) is the same as for the conservative mixing scale [60, 66], which is consistent with
 424 the fact that reactions are too slow to affect the progression of the front in this regime. Qualitatively, the
 425 first term depicts the compression of the reactive mixing scale due to the axial stretching and the associated
 426 perpendicular compression. On the other hand, the second term depicts the increase in the reactive mixing
 427 scale due to diffusion broadening at times where the reaction is occurring. The balance of these two terms
 428 is reached at the mixing time [52, 54]. Before the mixing time, compression is expected to dominate over
 429 diffusion, while the opposite situation develops after the mixing time for linear shear flows. The mixing

430 time can be estimated by recalling that we must have, at the transition, the balance between the width
 431 compression (the first term on the right hand side of equation (50)) and the diffusive growth (the second
 432 term on the right hand side of equation (50)). In the compression regime, the width of the reaction zone
 433 is given by the solution of $\frac{1}{s_r} \frac{\partial s_r}{\partial t} \sim \frac{1}{\delta} \frac{d\delta}{dt}$. which trivially yields the solution as $s_r \sim \delta$. We may thus write
 434 $\frac{1}{\delta} \frac{d\delta}{dt} \sim \frac{1}{2\delta^2}$ (where we have utilized the fact that $s_r \sim \delta$ at the transition). Simplifying this, we obtain the
 435 mixing time as $t_m \sim Pe^{-2/3}$ [60].

436 In summary, in the compression regime, $Pe^{-1} < t < Pe^{-2/3}$, the reactive mixing scale is expected to
 437 evolve by compression as $s_r \sim \frac{s_0}{\sqrt{1+Pe^2 t^2}}$. In the diffusion regime, $Pe^{-2/3} < t < Da^{-1}$, it is expected to grow
 438 diffusively as $s_r \sim \sqrt{t}$. For $t > Da^{-1}$, the progression of the reactive mixing scale is expected to be impeded
 439 by the consumption of reactants as described in the previous section and the mixing scale is expected to
 440 grow as $s_r \sim t^{1/6}$.

441 4.3. Reactive mixing scale dynamics with imposed flow: weak stretching, $Da^{-1} < Pe^{-1}$

442 We now consider the weak stretching scenario, for which the reaction time scale t_{Da_1} is reached earlier
 443 than the characteristic stretching time scale, t_{Pe_1} . To derive the temporal scaling of the reactive mixing
 444 scale in this regime, we appeal to the long time solution of equation (36), which can be expressed as,

$$445 \quad g \sim f(t) Ai \left(\frac{x' Da^{1/3}}{\delta(1 + Pe^2 t^2)^{1/3} \sqrt{\pi \theta^{1/3}}} \right). \quad (51)$$

446 where $f(t)$ is a function that depends only on time. Similarly to equation (31), this leads to a general form
 447 of the reaction rate, such as,

$$448 \quad R = Da \frac{x'}{t^{1/2}} g \sim h(t) \frac{x'}{s_r} Ai \left(C \frac{x'}{s_r} \right) \quad (52)$$

449 where $h(t)$ is a function that depends only on time, C is a constant, so that the functional form of the
 448 reaction width may be written as

$$449 \quad s_r \sim \delta(1 + Pe^2 t^2)^{1/3} \theta^{1/6} \quad (53)$$

449 Upon taking the log and differentiating equation (53), we obtain

$$450 \quad \frac{1}{s_r} \frac{\partial s_r}{\partial t} = \frac{1}{\delta} \frac{\partial \delta}{\partial t} - \frac{2}{3} \frac{Pe^2 t}{(1 + Pe^2 t^2)} + \frac{1}{6\theta} \frac{d\theta}{dt} \quad (54)$$

450 Thus, noting that $\frac{1}{\delta} \frac{d\delta}{dt} = \frac{Pe^2 t}{1+Pe^2 t^2}$, $\frac{d\theta}{dt} = 1/\delta^2$ and writing θ as a function of s_r through equation (53), we
 451 obtain the compression-diffusion equation in the case of weak stretching as

$$452 \quad \frac{1}{s_r} \frac{\partial s_r}{\partial t} = \frac{1}{3\delta} \frac{\partial \delta}{\partial t} + \frac{(1 + Pe^2 t^2)^{2/3}}{6(t + 1/3 Pe^2 t^3)^{2/3} s_r^2} \quad (55)$$

452 The compressive term on the right hand side of this equation is weak as compared to the diffusive expansion
 453 (as seen from the prefactor of $\frac{1}{3}$ in equation (55)) which is absent in equation (50).

454 We must also reiterate that the derivation shown above is strictly speaking true for the long time be-
 455 haviour (since we have appealed to the long time Airy function behavior for the concentration perturbation).
 456 This leads asymptotically to the $s_r \sim t^{1/6}$ behavior. Moreover, we shall see through the numerical results
 457 that at early times, the reaction width is completely governed by the compression term $\frac{1}{\delta} \frac{d\delta}{dt}$ seen in equa-
 458 tion (50). Essentially it means that the first term appearing through the long time analysis scenario of the
 459 compression-diffusion equation (55) does not play any role in determining the reaction mixing scale. Rather,
 460 in the initial moments, it is simply governed by the differential equation

$$\frac{1}{s_r} \frac{\partial s_r}{\partial t} = \frac{1}{\delta} \frac{\partial \delta}{\partial t} \quad (56)$$

461 For completeness, we still attempt to evaluate the mixing time for this scenario for which $\frac{1}{3\delta} \frac{\partial \delta}{\partial t} \sim$
 462 $\frac{(1+\text{Pe}^2 t^2)^{2/3}}{6\theta^{2/3} s_r^2}$. Upon substituting the form of $s_r \sim \delta^{1/3}$ in the compression regime and equating these two
 463 terms we obtain $t_m \sim \text{Pe}^{-2/3}$, which is the same as for the strong stretching regime.

464 The expressions derived here provide governing equations for the reactive mixing scale that allow quan-
 465 tifying the spatial localization of chemically reactive zone across mixing fronts. While equation (50) is the
 466 same as for the conservative mixing scale, equation (55) is different, which quantifies the limitation of the
 467 growth of the mixing scale by the consumption of reactant through chemical reactions. Regardless of the
 468 regimes described in section 4.3 and 4.2, the mixing time is always equal to $\text{Pe}^{-2/3}$, which is the same as for
 469 conservative mixing.

470 The dilution index quantifies the rate of mixing of a conservative scalar and has been utilized in recent
 471 times for the quantification of shear-induced-dilution [79]. In this regard, we may utilize the results obtained
 472 from the lamella approach to determine the dilution index. In Appendix C, we have depicted how the
 473 principles described herein may be employed to do so. As is proven, the minimum dilution occurs at a
 474 mixing time that is identical to that obtained in the preceding paragraph.

475 5. Numerical simulation of equations in the Lagrangian frame

476 In order to test the analytical expressions derived for the mass of product formed and the reactive mixing
 477 scale, we solve the set of equations (13) by means of a Chebyshev spectral collocation method [92]. This
 478 methodology has been employed in recent times to address a large assortment of nonlinear PDEs. Towards
 479 its implementation, we have to note that the domain is rescaled by L , where L is chosen sufficiently large over
 480 the desired observation time so as to prevent the boundary effects to affect the system evolution for the time
 481 span of interest. We denote the rescaled spatial variable as $\hat{z} = z/L$. The governing equations are subjected
 482 to the initial condition for a as $a(\hat{z}, 0) = 1 \ \forall \hat{z} \in (0, 1)$ and 0 otherwise. The initial condition for b is given
 483 by $b(\hat{z}, 0) = 1 - a(\hat{z}, 0)$ while for the species c we have the initial condition as $c(\hat{z}, 0) = 0$ everywhere. The
 484 boundary conditions are $a(1, \theta) = 0$, $a(-1, \theta) = 1$, $b(1, \theta) = 1$, $b(-1, \theta) = 0$ and $c(\pm 1, \theta) = 0$. The method

485 involves discretizing the 1D domain into the *Gauss-Lobatto* discretized grids represented by $\hat{z}_i = \cos(i\pi/N)$,
 486 where N represents the total number of intervals chosen for the solution. We make use of the Chebyshev
 487 differentiation matrices to represent the spatial derivatives. In a time discrete form (with the superscript k
 488 denoting the k^{th} time level), the discretized equation for the species a may be represented as

$$\frac{a^{k+1} - a^k}{\Delta\theta} = [a_{xx}]^{k+1} - \text{Da } a^k b^k (\delta^k)^2 \quad (57)$$

489 We may obtain the time-discrete equations for the evolution of species b and c along similar lines. Briefly
 490 speaking, the governing equations for the three species are expanded in the form of n Chebyshev polynomials
 491 of the first kind over the discretized domain \hat{z} as

$$a(\hat{z}) \approx \sum_{l=0}^n A_l T_l(\hat{z}) \quad (58)$$

492 We may write the discretized form of the reaction-diffusion equations as

$$\begin{aligned} a_i^{k+1} + \Delta\theta [a_{xx}]_i^{k+1} &= a_i^k - \text{Da } a_i^k b_i^k \frac{1}{1 + \text{Pe}^2(t^{k+1})^2} \\ b_i^{k+1} + \Delta\theta [b_{xx}]_i^{k+1} &= b_i^k - \text{Da } a_i^k b_i^k \frac{1}{1 + \text{Pe}^2(t^{k+1})^2} \\ c_i^{k+1} + \Delta\theta [c_{xx}]_i^{k+1} &= c_i^k + \text{Da } a_i^k b_i^k \frac{1}{1 + \text{Pe}^2(t^{k+1})^2} \end{aligned} \quad (59)$$

493 where the subscript i represents the value of the variable at the $i - th$ node while the superscript k denotes
 494 the $k - th$ time step. The initial and boundary conditions for equation (59) may be written as

$$\begin{aligned} a_i^0 &= 1 \quad \forall x > 0, b_i^0 = 1 - a_i^0, c_i^0 = 0 \\ a_N^k &= 1, a_0^k = 0, b_N^k = 1, b_0^k = 0, c_N^k = 0, c_0^k = 0 \quad . \end{aligned} \quad (60)$$

495 The spatial derivatives are represented by means of the Chebyshev differentiation matrices, \mathcal{D} , with $(\mathcal{D} a)$
 496 representing the first derivative of the vector a , $(\mathcal{D}^2 a)$ representing the second derivative of the vector a and
 497 so on. The boundary conditions of the discretized domains are incorporated by altering the first and last rows
 498 of the pertinent differentiation matrix, respectively. The MATLAB files for obtaining the spatiotemporal
 499 evolution of the concentration profiles and other derived parameters can be made available upon request.
 500 In the appendix we have demonstrated the validity of the numerical methodology presented here against
 501 full 2D simulations performed in the finite element framework of COMSOL Multiphysics. It may be seen
 502 from the figures that the simulations are in excellent agreement. The methodology presented here requires
 503 modest computational resources in comparison to the full 2D simulations [92]. In the following, we use this
 504 1D numerical model of diffusion, compression and reaction in the direction transverse to the front to explore
 505 the space of Pe and Da and validate the analytical expressions derived in the previous section.

506 6. Discussion

507 In this section we shall attempt to shed light on the temporal scaling of the mass produced and trends
508 observed for the transition times among the different regimes as obtained from the theoretical discussion in
509 this work, and compare it against the numerical results obtained by means of the 1D Chebyshev spectral
510 collocation method. We shall first focus on the cases of no imposed flow and imposed flow (with the two
511 subcases of weak stretching and strong stretching). We then proceed to study the temporal evolution of the
512 reactive mixing scale for the cases of no shear and shear while attempting to quantify the observed temporal
513 evolutions by means of the compression-diffusion equation proposed in section 4.

514 6.1. Temporal evolution of the mass of product

515 6.1.1. No shear scenario

516 For completeness, we first depict the temporal evolution of the mass of product for the case where there
517 is no imposed flow. In figure 3(a) we depict the temporal variation of the mass of the product formed for
518 different Damköhler numbers ($\text{Da} = 0.1, 1, 10, 100$). We observe from figure 3(a) that the initial scaling
519 obtained from the numerical simulation does corroborate excellently with the theoretical predictions made
520 in section 3.1.1; in the initial reaction-diffusion regime (reaction kinetics dominated regime), the mass scales
521 as $m_c \sim \text{Da}t^{3/2}$, as predicted analytically. Essentially we observe that as Da increases, there is a concomitant
522 increase in the product mass produced. Beyond the transition time $t_{\text{Da}_1} = 32/\text{Da}$ the mass scaling is no more
523 limited by reaction kinetics and evolves as per the scaling $m_c \sim t^{1/2}$, corresponding to the diffusion-limited
524 regime, as theoretically obtained in section 3.1.2. In accordance to the theoretical predictions, it is seen from
525 figure 3(a) that beyond the time t_{Da_1} , m_c becomes independent of the Damköhler number. Clearly, this
526 underpins the fact that beyond the transition time, the evolution is limited by diffusion and not dictated
527 by the reaction kinetics term. In figure 3(b), we depict the temporal evolution of the rescaled mass of the
528 product $m_c/m_{c,\text{Da}_1}$ as a function of the rescaled time t/t_{Da_1} . The curves depicted in figure 3(a) collapse on
529 top of each other, thus confirming the validity of the theoretical predictions of the rescaling of the transition
530 time and the mass of the product formed at that particular transition time.

531 6.1.2. Weak stretching scenario

532 We now proceed to analyze the weak stretching scenario $\text{Da}^{-1} < \text{Pe}^{-1}$. In figure 4 we depict the
533 temporal evolution of the mass of the product formed for $\text{Pe} = 0.1$ and $\text{Da} = 0.1, 1, 10, 100$. Initially all
534 the curves increase as per the scaling $m_c \sim t^{3/2}$, indicating the kinetics-limited regime. After this initial
535 scaling, depending on the magnitude of Da , a transition from the $m_c \sim t^{3/2}$ scaling to the $m_c \sim t^{1/2}$ scaling
536 is observed. The dependence of the transition time on Da is consistent with the predicted relationship
537 $t \sim \text{Da}^{-1}$. Then at around $t \sim \text{Pe}^{-1}$ all the curves merge together to follow a mass scaling in the form $t^{3/2}$,
538 which is consistent with the expected shear-enhanced reactive mixing regime. For the case $\text{Da} = \text{Pe} = 0.1$,

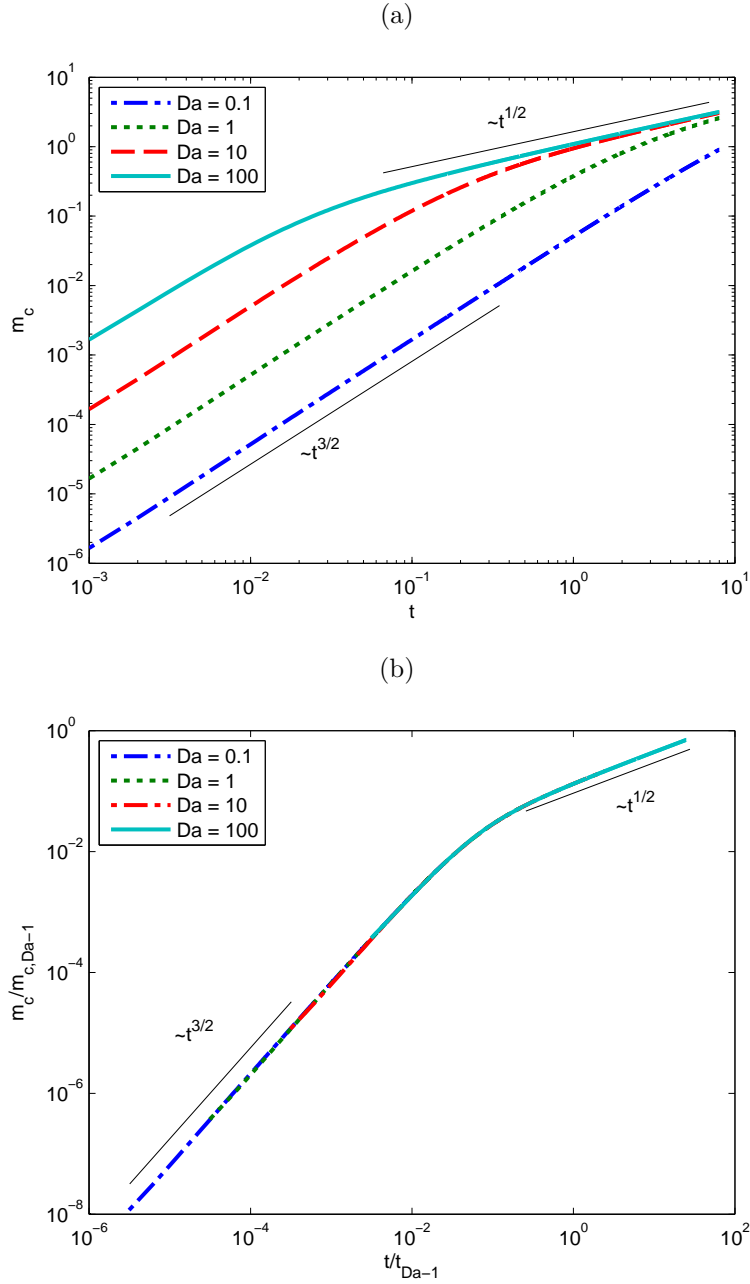


Figure 3: (a) Temporal evolution of the mass of product formed for $Da = 0.1, 1, 10, 100$ in the case where $Pe = 0$, as obtained from the Chebyshev spectral method. (b) Rescaled temporal evolution of the mass of product formed for $Da = 0.1, 1, 10, 100$ for the case where there is no externally imposed flow. Towards rescaling the aforementioned variables, the time has been rescaled as $t/(32/Da)$ while the mass has been rescaled as $m_c/(45/\sqrt{Da})$.

539 the intermediate regime does not exist and the system transits directly from the kinetics limited regime to
 540 the long-time shear-enhanced reactive mixing regime (wherein both the regimes have a temporal scaling of
 541 $m_c \sim t^{3/2}$). Besides this, we also notice that, in accordance to the theoretical prediction, the long time

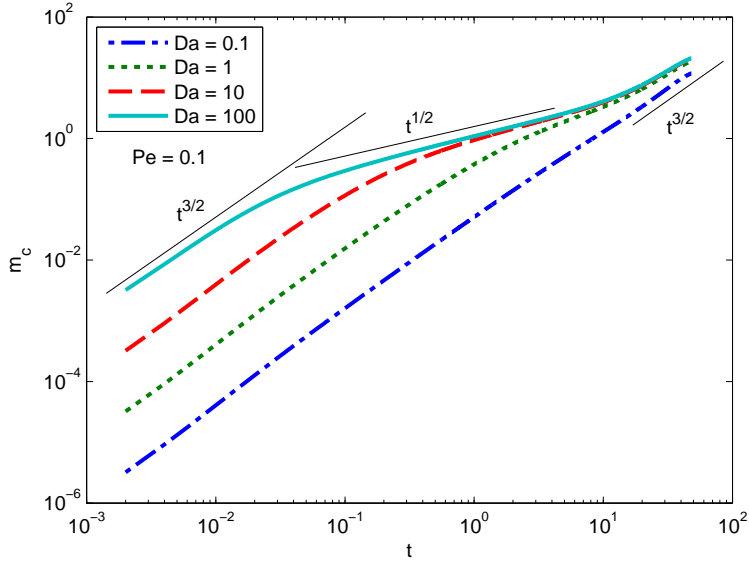


Figure 4: Temporal evolution of the product mass for $Pe = 0.1$ for $Da = 0.1, 1, 10, 100$. The three distinct regimes, reaction controlled, diffusion controlled and stretching controlled, are observed when the two transition times are separated (i.e. $Pe < Da$). As Pe becomes comparable to Da , the intermediate regime of diffusion-limited growth is not observed.

542 behavior of m_c is independent of Da . Upon rescaling the curves of figure 4 by the first transition time
 543 $t_{Da_1} = 32/Da$ for the time axis, and the mass of product at this time $m_{c,Da_1} \approx 45/\sqrt{Da}$, they collapse into
 544 each other (figure 5(a)) for the first two regimes (i.e. for $t < t_{Pe_1}$). This confirms the validity of the predicted
 545 scalings for the first transition time t_{Da_1} . In figure 5(b) we observe that the curves for different Pe (and
 546 different Da as well) collapse onto each other for the later two regimes (i.e. for $t > t_{Da_1}$) when we rescale
 547 time for the curves in figure 4 by the second transition time $t_{Pe_1} \approx Pe^{-1}$ and the mass of the product
 548 formed by $m_c \approx 8Pe^{-1/2}$.

549 We remark that all the observations are in complete agreement with the theoretical predictions presented
 550 in section 3. The regime $Da^{-1} < Pe^{-1}$ thus essentially decouples the effect of stretching, active for $t > Pe^{-1}$,
 551 from the effect of kinetics limitations, which are significant for $t < Da^{-1}$. In what follows, we shall consider
 552 the case where the fluid stretching is coupled to the chemical reactions, i.e. the strong stretching scenario
 553 where $Pe^{-1} < Da^{-1}$.

554 6.1.3. Strong stretching scenario

555 In figure 6(a) we investigate the strong stretching scenario $Pe^{-1} < Da^{-1}$. We consider the temporal
 556 evolution of m_c for the cases where $Pe = 100$ and $Da = 0.1, 1, 10, 100$. For $t < Pe^{-1}$, we observe that
 557 the evolution of m_c scales as $m_c \sim t^{3/2}$. After this dimensionless time, stretching becomes significant
 558 while reaction kinetics is still a limiting factor since $t < Da^{-1}$. Hence, in this intermediate regime we

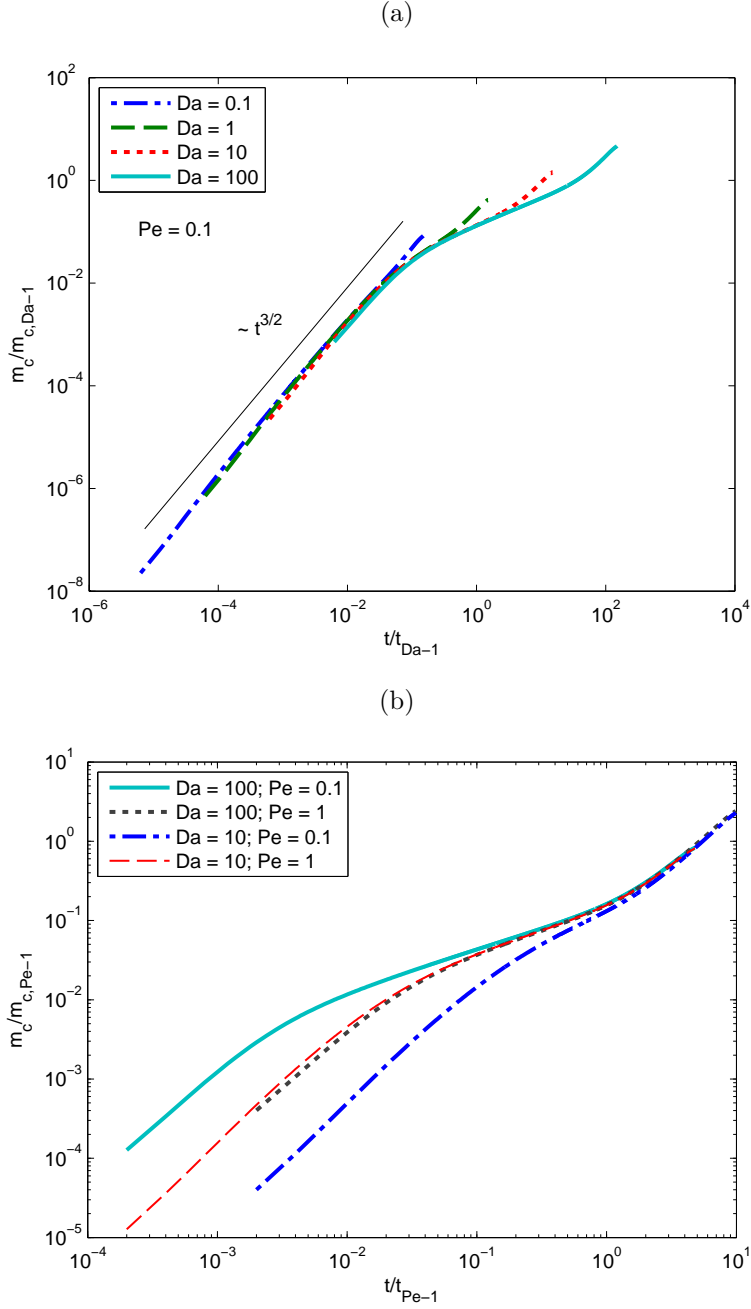


Figure 5: (a) Scaled mass of product vs time $m_c/m_{c, Da_1}$ as a function of t/t_{Da_1} , for $Pe = 0.1$ and $Da = 0.1, 1, 10, 100$ at the first transition time for weak stretching, t_{Da_1} . (b) Scaled mass of product vs time, for $Pe = 0.1$ and $Pe = 1$ for $Da = 10, 100$ at the second transition time for weak stretching, t_{Pe_1} .

559 observe the predicted accelerated scaling $m_c \sim t^{5/2}$, induced by the coupling between kinetics limitation
 560 and stretching. This first transition is confirmed by rescaling time by $t_{Pe_2} = 5\sqrt{3}Pe^{-1}/2$ and mass by
 561 $m_{c, Pe_2} = (5\sqrt{3}/2)^{3/2}/Pe^{3/2}$, which collapses the curves together (please refer to figure 7 (a)). The second

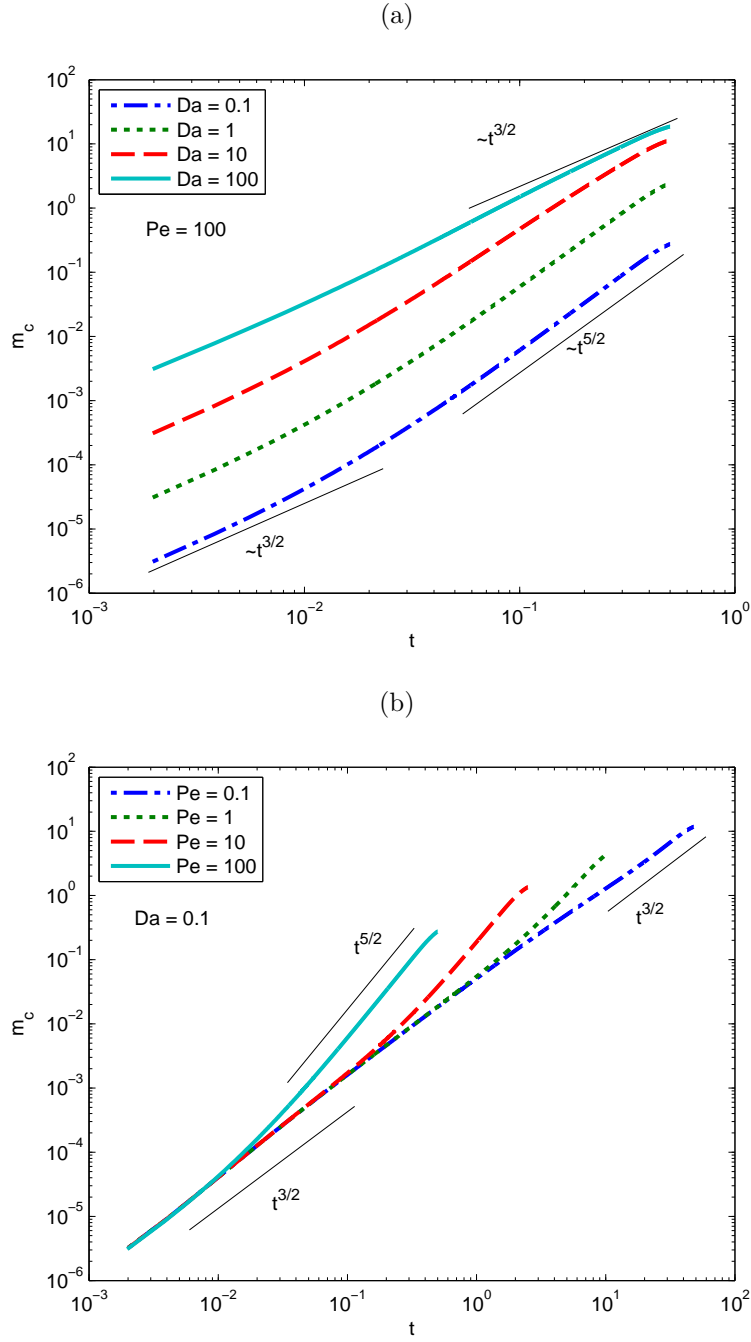


Figure 6: (a) Temporal evolution of the product mass for $Pe = 100$ and $Da = 0.1, 1, 10, 100$. As opposed to the case considered in figure 4, we consider the effect of strong stretching, thus allowing the system to undergo the transition into the stretching controlled regime at early times. (b) Temporal evolution of the product mass for $Da = 0.1$ and $Pe = 100, 10, 1, 0.1$.

562 transition from $m_c \sim t^{5/2}$ regime to $m_c \sim t^{3/2}$ regime is not visible on a single simulation due to very
 563 long time of simulation required. This transition is however visible when collapsing the different simulations

564 together through rescaling time by $t_{\text{Da}_2} \approx 80\sqrt{3}/\text{Da}$ and mass by $m_{c,\text{Da}_2} \approx 8(80\sqrt{3})^{3/2}\text{Pe}/\text{Da}^{3/2}$ (figure
565 7(b)).

566 Note that the intermediate scaling is not observed for $\text{Pe} = \text{Da} = 100$, since the system transitions directly
567 from the early time kinetics-limited regime with no significant stretching to the stretching enhanced reactive
568 mixing regime with no kinetics limitation, which both scale as $m_c \sim t^{3/2}$. The agreement with theoretical
569 derivations is further confirmed by the series of simulations performed for $\text{Da} = 0.1$ and $\text{Pe} = 0.1, 1, 10, 100$
570 (figure 6(b)). The transition t_{Pe_2} is readily seen in this case when considering the trends of the transition
571 from the $m_c \sim t^{3/2}$ regime to the $m_c \sim t^{5/2}$ regime as Pe is increased. For the situation for which $\text{Da} \sim \text{Pe}$,
572 we see again that the region in which there is a coupling of the reaction kinetics and stretching vanishes as
573 expected. Figure 7(a) clearly depicts that when the time and the mass of the product are rescaled by t_{Pe_2}
574 and m_{c,Pe_2} respectively, we observe that the curves in figure 6(b) collapse onto each other for the first two
575 regimes (i.e. for $t < \text{Da}^{-1}$). On the other hand figure 7(b) clearly depicts that when the time and the mass
576 of the product are rescaled by t_{Da_2} and m_{c,Da_2} , we obtain a good collapse of the curves depicted in figure
577 6(a) for the other later two regimes (i.e. for $t > \text{Pe}^{-1}$).

578 6.2. Temporal evolution of the reactive mixing scale

579 In figure 8 we depict the temporal evolution of the reactive mixing scale, s_r as a function of time for
580 configurations of strong stretching as manifested through a low Damköhler ($\text{Da} = 0.1$) and a Péclet that
581 is larger than the Damköhler ($\text{Pe} = 0.1, 1, 10, 100$). We observe a good agreement with the compression-
582 diffusion equation elucidated in section 4.2. There is an initial diffusion controlled growth of the reactive
583 mixing scale until the time Pe^{-1} . After this time we observe a compression (the reactive mixing scale
584 decreases), followed by a transition at $t \sim \text{Pe}^{-2/3}$ to the $s_r \sim t^{1/2}$ regime.

585 In figure 9 we depict the evolution of the reactive mixing scale as a function of time for configurations
586 of weak stretching with $\text{Da} = 100$ and $\text{Pe} = 0.1, 1, 10, 100$. It may be observed from figure 9 that at small
587 times the reactive mixing scale grows at a rate which is limited by diffusion until the time $t \sim \text{Pe}^{-1}$ at
588 which a compression occurs in the system. This is most prominently seen in the case of $\text{Pe} = 100$ and
589 $\text{Pe} = 10$. In between the stretching time Pe^{-1} and the mixing time $\text{Pe}^{-2/3}$ we observe that the impact
590 of the stretching is to compress the reactive mixing scale. This compression occurs until the mixing time
591 $t_m \sim \text{Pe}^{-2/3}$, beyond which we observe a growth of the reactive mixing scale as $s_r \sim t^{1/6}$. The latter
592 is expected since the dimensionless time is larger than the Da^{-1} and hence the expansion of the reaction
593 mixing scale is impeded by the rate of consumption of the reactants. The solution of equation (55) is seen
594 to be valid after the reactive mixing time. However it is unable to predict the observed compression. This
595 may be attributed to the fact that equation (55) is derived by taking into consideration the scaling form
596 at large times (please refer to section 4.3). It is however observed that the compression-diffusion equation
597 (equation (50), derived in section 4.2) is able to predict the early time behavior for the curves to a good

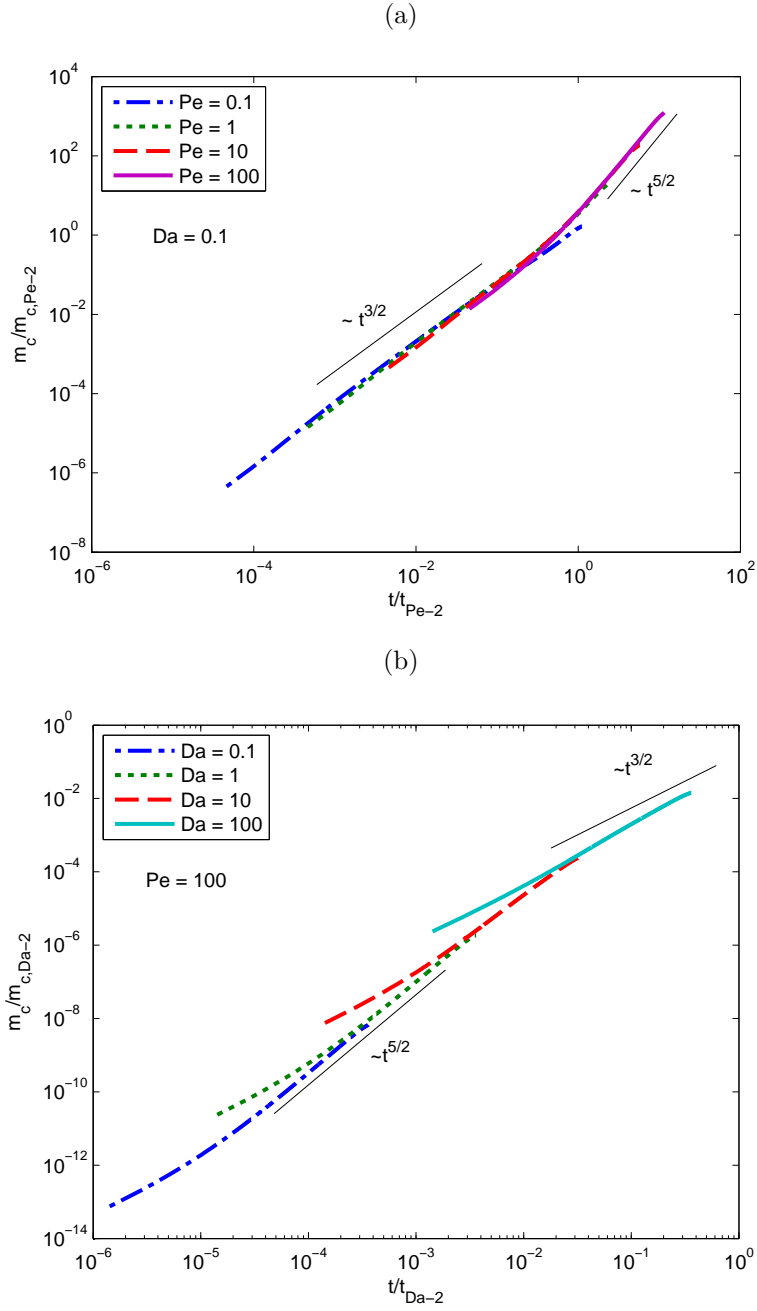


Figure 7: (a) Rescaled mass of product, $m_c/m_{c,Pe_2}$, vs rescaled time, t/t_{Pe_2} , for $Da = 0.1$ (strong stretching) and $Pe = 0.1, 1, 10, 100$ at the first transition time, t_{Pe_2} . (b) Rescaled mass of product, $m_c/m_{c,Da_2}$, vs rescaled time, t/t_{Da_2} , for $Pe = 100$ (strong stretching) and $Da = 0.1, 1, 10, 100$ at the second transition time, t_{Da_2} .

598 approximation. Consequently, this observation suggests that the chemical reactions do not affect the early
 599 time behavior of the reactive mixing scale.

600 Moving beyond the extreme cases of Pe and Da in figure 10 we represent the temporal evolution of the

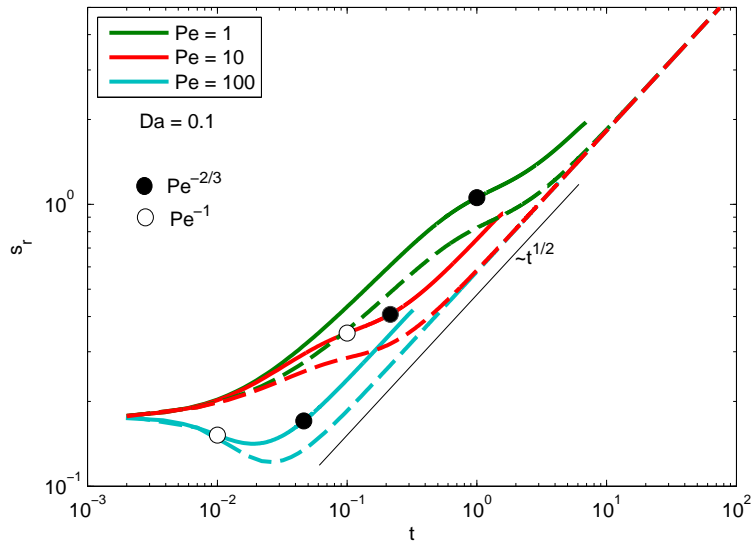


Figure 8: Temporal evolution of the reactive mixing scale for a case for which Pe is larger than Da . We have chosen $Da = 0.1$ and depicted the reactive mixing scales for $Pe = 1, 10, 100$. The black filled markers corresponds to the mixing time $t_m = Pe^{-2/3}$ while the white filled marker corresponds to the time Pe^{-1} for $Pe = 10$ and 100 . The solid lines represent the solution from the 1D Chebyshev spectral collocation method while the dashed lines represent the prediction from the compression diffusion equation (50)

601 reactive mixing scale for a situation where Pe and Da are moderate and not much different from each other
 602 in a hope to outline the transition from one kind of behavior to the other kind of behavior. For this case we
 603 have chosen $Da = 10$ and $Pe = 1$. The solid line represents the numerical solution while the dash-dot line
 604 represents the solution obtained from equation (50) while the dashed line represents the solution obtained
 605 from equation (55). It may be clearly observed that the initial evolution of the reactive mixing scale is
 606 reasonably predicted by equation (50) while beyond the overlap of the two curves, the curve is depicted in
 607 a better fashion by (55). The crossover in behavior occurs at the the mixing time scale, which in this case
 608 is close to 1.

609 7. Conclusions

610 This study provides analytical expressions of upscaled effective reaction rates in the presence of shear
 611 flow. These expressions quantify the coupling between mixing limitations, governed by fluid stretching and
 612 diffusion and quantified by the Péclet number, and kinetic limitations, quantified by the Damköhler number.
 613 It is seen that velocity gradients promote mixing by elongating the interface available for diffusive mass
 614 transfer and increasing concentration gradients. As discussed in this study, the coupling of this phenomenon
 615 with a broad range of characteristic reaction time scales is non-trivial. We have thus quantified the impact

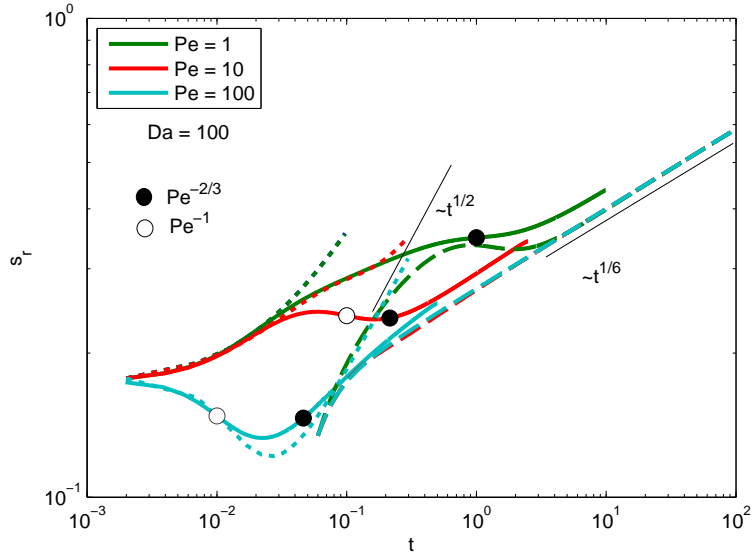


Figure 9: Temporal evolution of the reactive mixing scale as a function of time for $Da = 100$ and $Pe = 1, 10, 100$. The black filled markers correspond to the mixing time $t \sim Pe^{-2/3}$ while the white filled marker correspond to the time Pe^{-1} for the cases $Pe = 10$ and 100 . The solid lines represent the numerical solution obtained using the 1D Chebyshev spectral collocation method, the dotted lines represent the solution obtained from the compression-diffusion equation in section 4.2, and the dashed curves represent the curves obtained using the compression-diffusion equation in section 4.3.

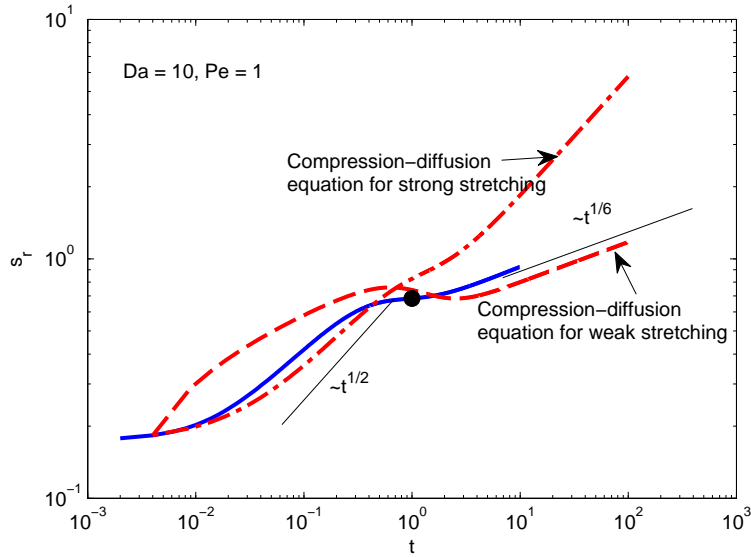


Figure 10: Temporal evolution of the reactive mixing scale as a function of time for the case with $Da = 10$ and $Pe = 1$. The solid line represents the solution obtained from the 1D Chebyshev spectral collocation method. The dashed and the dash-dot-dash line represent the solutions from the compression diffusion equations (55) and (50), respectively.

616 of the Damköhler and Péclet numbers on the effective reaction kinetics of mixing front subject to shear
617 deformation. Different regimes have been identified theoretically in this temporal behavior, as well as the
618 transition times between them. Through the functional dependency of the reactive mixing scale on time, we
619 have analyzed the spatial localization of reactivity as a function of time, Damköhler and Péclet numbers.
620 We have also proposed an efficient and fast numerical scheme based on a Chebyshev spectral collocation
621 method in order to tackle the nonlinear reaction-advection-diffusion problem numerically, which has allowed
622 us to verify successfully all our theoretical predictions.

623 While previous works had mostly focused on the cases of $Pe = 0$ with variable Da or $Da = \infty$ with
624 variable Pe , the theoretical framework presented here spans the full space of Pe and Da (see figure 2). In
625 the case of a stretching that is weak in comparison to the reaction, i.e. for $Pe < Da$, the effects of kinetics
626 limitations and mixing enhancement by shear are essentially decoupled. Kinetics limitation is dominant
627 in the early time regime for $t < t_{Da_1} = 32/Da$, while stretching enhanced mixing is dominant in the late
628 time regime $t > t_{Pe_1} = 1/Pe$. In the intermediate regime reactions are limited by diffusion, leading to the
629 classical diffusive scaling. In the case of a stretching that is strong in comparison to the reaction kinetics
630 , i.e. for $Pe > Da$, we have shown the existence of an intermediate regime at times $t_{Pe_2} < t < t_{Da_2}$, with
631 $t_{Pe_2} = \frac{1}{Pe}5\sqrt{3}/2$ and $t_{Da_1} = \frac{1}{Da}80\sqrt{3}$, where stretching enhanced mixing and kinetics limitations are strongly
632 coupled. This leads to an accelerated effective kinetics with $m_c \approx \frac{PeDa}{10\sqrt{3}}t^{5/2}$.

633 The presence of a background velocity gradient affects the evolution of the reactive mixing scale, which
634 defines the region around the interface between the two reactants where reaction mostly occurs. In the
635 absence of an imposed shear, the reactive mixing scale initially grows as $s_r \sim t^{1/2}$, due to control by
636 molecular diffusion. Beyond the time t_{Da_1} this growth slows down as $s_r \sim t^{1/6}$ due to consumption of
637 reactants by the chemical reaction. The presence of a velocity gradient affects this evolution by introduction
638 a regime of compression in the temporal evolution of the reactive mixing scale. We have attempted to
639 formalize the nature of this evolution of the reactive mixing scale by introducing a compression-diffusion
640 equation for each of the two different cases of weak and strong stretching. We have shown that the reactive
641 layer is compressed between the characteristic stretching time $t \sim Pe^{-1}$ and the mixing time $t_m \sim Pe^{-2/3}$, at
642 which diffusion balances compression. Beyond this time the reactive mixing scale evolves as $s_r \sim t^{1/6}$ in the
643 case of weak stretching and $s_r \sim t^{1/2}$ in the case of strong stretching. The compression-diffusion equations
644 are able to predict the nature of the evolution of the reactive mixing scale in their respective domains of
645 validity with reasonable accuracy.

646 The results presented in this work represent a step towards understanding how complex stretching
647 processes found in particular in porous media interact with the reactivity of transported solutes to determine
648 the dynamics of upscaled effective kinetics and the degree of spatial localization of reactive hotspots. While
649 the presented results are directly relevant to 2D porous media where linear deformation has been observed
650 [81], they may also be extended to power law deformation [66] and exponential deformation [80]. Such

651 stretching dynamics may be related to topological metrics such as the Okubo-Weiss (OW) parameter [79].
652 The presented methodology, developed here for a constant velocity gradients, may be extended to spatially
653 variable velocity gradients based on the lamella representation of mixing fronts. For any given flow field,
654 we may always consider a stretched front to be comprised of a large number of small elements [54]. In
655 this way, locally, we may address the evolution of the concentration of the species involved in the direction
656 perpendicular to the strip (the local moving coordinate system comprises of two directions: one direction
657 points along the lamella while the other is perpendicular to it). Based on the ideas in this work, we may
658 thus extend this framework to account for reactions between two species encountering arbitrary flow fields.

659 However, we must briefly mention the drawbacks of the reactive lamella approach and accordingly men-
660 tion a prospective road map for potential research. Since chemical reactions introduce strong non-linearities,
661 we assumed that a given isolated lamella does not interact with other lamellae (note that this is not an issue
662 for conservative mixing processes since lamella concentrations add up linearly upon overlapping). While
663 this hypothesis is satisfied exactly for stratified flows, the same may not hold true for other flow fields, for
664 example in situations where there is a severe bending of the material line which renders the evolution of
665 the concentration fields in that local coordinate system to be necessarily 2D instead of 1D. For such flows,
666 the lamella approach is expected to work best for the case of high Péclet numbers (wherein the diffusion
667 is small compared to advection). We have assumed that the diffusion coefficients of all the species in the
668 reaction are equal. This assumption allows us to make several analytical treatments to the reduced set of
669 governing equations in the warped coordinates and obtain underlying scaling regimes and transition times
670 in terms of the Péclet and Damköhler numbers. A more general formulation to study the reaction dynamics
671 of such a system would entail relaxation of this assumption [45, 46]. In the situation where the diffusion
672 coefficients are different, one would obtain travelling fronts which exhibit a rich range of dynamics. This
673 can potentially form the basis for further development based on this work towards identifying the impact
674 of diffusivity contrast on enhanced reaction kinetics in the presence of shear flows. Going beyond 2D, these
675 ideas may be extended to 3D flows, which are inevitable when considering groundwater flow. The evolution
676 of material volumes in 3D can occur either in the form of sheets or fingers, depending on the local flow
677 structure (see [55] for several such situations). For the former scenario, we may apply a similar procedure
678 for the sheets in which the direction normal to the sheets is the primary direction of species dynamics. For
679 the later scenario, however, we may conceptualize the fingers (or equivalently tube-like structures) in a local
680 cylindrical coordinate in which the axial direction is aligned along the direction of the strip and the radial
681 direction is the normal direction along which the species dynamics occurs. Interestingly, one possibility is
682 to use this framework in conjugation with information about the conductivity field of a porous media which
683 may be related to the temporal evolution of stretching for a given material line (cf. [93]). Thus, prospects
684 for further work include extending the numerical framework presented here to more complex stretching
685 configurations, involving spatial and temporal fluctuations in stretching rates, as well as more complicated

686 reaction kinetics including multistep or higher order reactions.

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691 Appendix A. Transformation into a moving coordinate system

We consider a linear transformation into the coordinate system moving and rotating with the material line,

$$\tilde{\mathbf{x}}' = \mathbf{A}^\top(t) [\tilde{\mathbf{x}} - \tilde{\mathbf{x}}(\tilde{t})], \quad \frac{d\tilde{\mathbf{x}}(\tilde{t})}{dt} = \mathbf{u}[\tilde{\mathbf{x}}(\tilde{t})] \quad (\text{A.1})$$

where $\mathbf{A}(\tilde{t})$ is an orthogonal matrix specified below in equation (A.11). Thus, concentration $\tilde{a}(\tilde{\mathbf{x}}, \tilde{t})$ is given in terms of the concentration $\tilde{a}'(\tilde{\mathbf{x}}, \tilde{t})$ in the moving coordinate system as

$$\tilde{a}(\tilde{\mathbf{x}}, \tilde{t}) = \tilde{a}'(\mathbf{A}^\top(\tilde{t}) [\tilde{\mathbf{x}} - \tilde{\mathbf{x}}(\tilde{t})], \tilde{t}), \quad (\text{A.2})$$

and correspondingly for $\tilde{b}(\tilde{\mathbf{x}}, \tilde{t})$ and $\tilde{c}(\tilde{\mathbf{x}}, \tilde{t})$. Inserting (A.2) into (1) for $\tilde{a}(\tilde{\mathbf{x}}, \tilde{t})$ gives

$$\begin{aligned} \frac{\partial \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t})}{\partial t} + \left[\frac{d\mathbf{A}^\top(\tilde{t})}{dt} \mathbf{A}(\tilde{t}) \tilde{\mathbf{x}}' - \mathbf{u}[\tilde{\mathbf{x}}(\tilde{t})] \mathbf{A}(\tilde{t}) \right] \nabla' \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) + \mathbf{u} [\mathbf{A}(\tilde{t}) \tilde{\mathbf{x}}' + \tilde{\mathbf{x}}(\tilde{t})] \mathbf{A}(\tilde{t}) \nabla' \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) = \\ D \nabla' \cdot \mathbf{A}^\top \mathbf{A}(\tilde{t}) \nabla' c'(\tilde{\mathbf{x}}', \tilde{t}) - k \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) b'(\tilde{\mathbf{x}}', \tilde{t}). \end{aligned} \quad (\text{A.3})$$

We note that $\mathbf{A}^\top(\tilde{t}) \mathbf{A}(\tilde{t}) = 1$ due to the orthogonality of \mathbf{A} . Furthermore, we expand

$$\mathbf{u} [\mathbf{A}(\tilde{t}) \tilde{\mathbf{x}}' + \tilde{\mathbf{x}}(\tilde{t})] = \mathbf{u} [\tilde{\mathbf{x}}(\tilde{t})] + \boldsymbol{\epsilon}(\tilde{t}) \mathbf{A}(\tilde{t}) \tilde{\mathbf{x}}' + \dots, \quad (\text{A.4})$$

where we have defined the deformation rate tensor

$$\epsilon_{ij}(\tilde{t}) = \frac{\partial u_i[\tilde{\mathbf{x}}(\tilde{t})]}{\partial \tilde{x}_j}. \quad (\text{A.5})$$

Inserting (A.4) into (A.3) gives

$$\frac{\partial \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t})}{\partial t} + \left[\frac{d\mathbf{A}^\top(\tilde{t})}{dt} \mathbf{A}(\tilde{t}) + \mathbf{A}^\top(\tilde{t}) \boldsymbol{\epsilon}(\tilde{t}) \mathbf{A}(\tilde{t}) \right] \tilde{\mathbf{x}}' \cdot \nabla' \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) = D \nabla'^2 \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) - k \tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) b'(\tilde{\mathbf{x}}', \tilde{t}). \quad (\text{A.6})$$

692 The expression in the square brackets is the deformation rate tensor $\boldsymbol{\epsilon}'(\tilde{t})$ in the moving coordinate system.

693 As pointed out in [55], it is a pseudotensor.

For the simple shear flow $\mathbf{u}(\tilde{\mathbf{x}}) = G y \mathbf{e}_x$ under consideration here, the deformation rate tensor is

$$\boldsymbol{\epsilon}(\tilde{t}) = \begin{pmatrix} 0 & G \\ 0 & 0 \end{pmatrix}. \quad (\text{A.7})$$

A material strip $\tilde{\ell}(\tilde{t})$ oriented initially perpendicular to the flow direction evolves as

$$\tilde{\ell}(\tilde{t}) = \tilde{\ell}_0 \begin{pmatrix} G\tilde{t} \\ 1 \end{pmatrix} \quad (\text{A.8})$$

where $\tilde{\ell}_0 = |\tilde{\ell}(t=0)|$ is the initial elongation. The strip length $\tilde{\ell}(\tilde{t}) = |\tilde{\ell}(\tilde{t})|$ is (please refer to figure 1)

$$\tilde{\ell}(\tilde{t}) = \tilde{\ell}_0 \sqrt{1 + (G\tilde{t})^2}. \quad (\text{A.9})$$

Due to volume conservation, $\tilde{\ell}(\tilde{t})\tilde{\delta}(\tilde{t}) = \tilde{\ell}_0\tilde{\delta}_0$, the width of the strip is given by

$$\tilde{\delta}(\tilde{t}) = \frac{\tilde{\delta}_0}{\sqrt{1 + (G\tilde{t})^2}}. \quad (\text{A.10})$$

We transform into the coordinate system whose x' -axis is oriented perpendicular to the material strip. Thus, the orthogonal matrix $\mathbf{A}(\tilde{t})$ is here given by

$$\mathbf{A}(\tilde{t}) = \frac{1}{\sqrt{1 + (G\tilde{t})^2}} \begin{pmatrix} 1 & G\tilde{t} \\ -G\tilde{t} & 1 \end{pmatrix} \quad (\text{A.11})$$

So, we obtain for $\epsilon'(\tilde{t})$ the triangular form

$$\epsilon'(\tilde{t}) = \frac{1}{1 + (G\tilde{t})^2} \begin{pmatrix} -G^2\tilde{t} & 0 \\ G - G^3\tilde{t}^2 & G^2\tilde{t} \end{pmatrix}. \quad (\text{A.12})$$

Initially, the concentration is uniformly distributed along the solute front, see (6), that is, in the moving coordinate system $\tilde{a}'(\tilde{\mathbf{x}}', \tilde{t} = 0) = 1 - H(\tilde{x}')$ and $\tilde{b}'(\tilde{\mathbf{x}}', \tilde{t} = 0) = H(\tilde{x}')$ with $H(\tilde{x}')$ the Heaviside step function. Due to symmetry, the species concentrations are independent of \tilde{y}' , this means $\tilde{a}'(\tilde{\mathbf{x}}', \tilde{t}) \equiv \tilde{a}'(\tilde{x}', \tilde{t})$ and $\tilde{b}'(\tilde{\mathbf{x}}', \tilde{t}) \equiv \tilde{b}'(\tilde{x}', \tilde{t})$. Thus, for the above initial condition, the advection-diffusion problem (A.6) across the strip simplifies to

$$\frac{\partial \tilde{a}'(\tilde{x}', \tilde{t})}{\partial \tilde{t}} - \Omega(\tilde{t})\tilde{x}' \frac{\partial \tilde{a}'(\tilde{x}', \tilde{t})}{\partial \tilde{x}'} - D \frac{\partial^2 \tilde{a}'(\tilde{x}', \tilde{t})}{\partial \tilde{x}'^2} = -k\tilde{a}'(\tilde{x}', \tilde{t})\tilde{b}'(\tilde{x}', \tilde{t}), \quad (\text{A.13})$$

where we have defined

$$\Omega(\tilde{t}) = \frac{G^2\tilde{t}}{1 + (G\tilde{t})^2} = \frac{1}{\tilde{\ell}(\tilde{t})} \frac{d\tilde{\ell}(\tilde{t})}{d\tilde{t}} = -\frac{1}{\tilde{\delta}(\tilde{t})} \frac{d\tilde{\delta}(\tilde{t})}{d\tilde{t}}. \quad (\text{A.14})$$

694 For simplicity of notation, we drop the primes for the remainder of the paper.

695 Appendix B. Validation of 1D methodology

696 We present here a validation of the 1D Chebyshev spectral collocation method which has been employed
697 in the present work. We compare the temporal evolution of the mass of the product formed against 2D
698 simulations performed in the finite element framework of COMSOL Multiphysics. For the 2D simulation, we

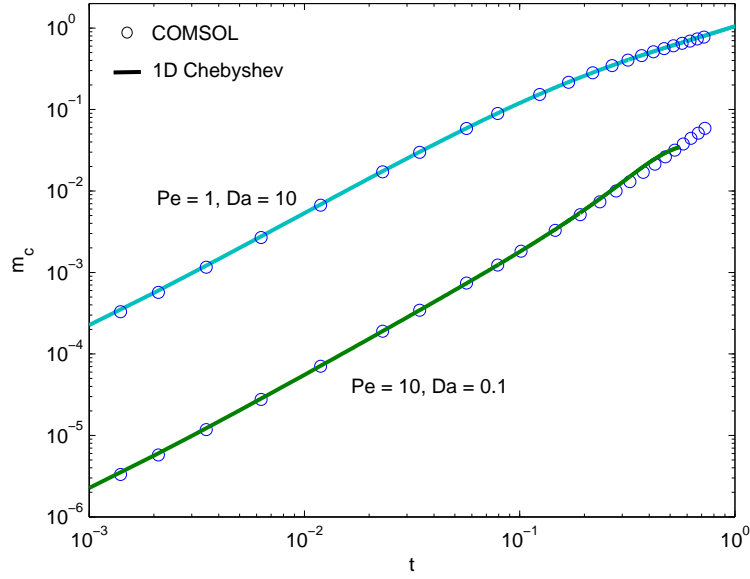


Figure B.11: Temporal evolution of the mass of product formed for two different cases: $Da = 0.1, Pe = 10$ and $Da = 10, Pe = 1$. The symbols denote the results obtained from the COMSOL 2D simulations, while the solid lines are those obtained from the 1D Chebyshev collocation method (please see 5).

699 consider the case of a stretching front for two different cases, $Da = 0.1, Pe = 10$ and $Da = 10, Pe = 1$. The
700 grid spacing employed in COMSOL is 0.1 while the domain size is taken to be 20×20 . Each front is chosen
701 to be of size 10, which, compared to the nondimensional length 1 is large enough to prevent the periodic
702 boundary condition from interacting with the diffusion front in the time of interest of our observation (we
703 refer the reader to section 2.2 for a discussion regarding this). The minimum grid size decides the initial
704 width of the reaction front and, consequently, the system evolution. For the 1D spectral simulation presented
705 in figure B.11, we have employed a domain length of 30 with $N = 384$ intervals for the 1D Chebyshev spectral
706 collocation method. The time step in the warped time system is chosen to be $\Delta\theta = 10^{-3}$.

707 If we focus on the periodic boundary condition employed, we may assume that the effects of diffusion in
708 the Lagrangian frame are primarily focused on the region near the interface. Only when the reactive mixing
709 scale becomes large enough to overlap with the reactive mixing scales of the products formed at the periodic
710 ends, does the assumption that $z \rightarrow \pm\infty$, which signifies the semi-infinite domain as per the 1D analysis
711 in the present work, begins to lose its validity. Our study focuses on the behavior of the system at times
712 before the merging of the two fronts happens.

713 It appears clearly in figure B.11 that the temporal evolution of the mass of the product, m_c , obtained
714 through the two aforementioned methods (the solution obtained through the 2D simulations in COMSOL
715 Multiphysics is denoted by the symbols while that obtained from the 1D methodology is denoted by the solid

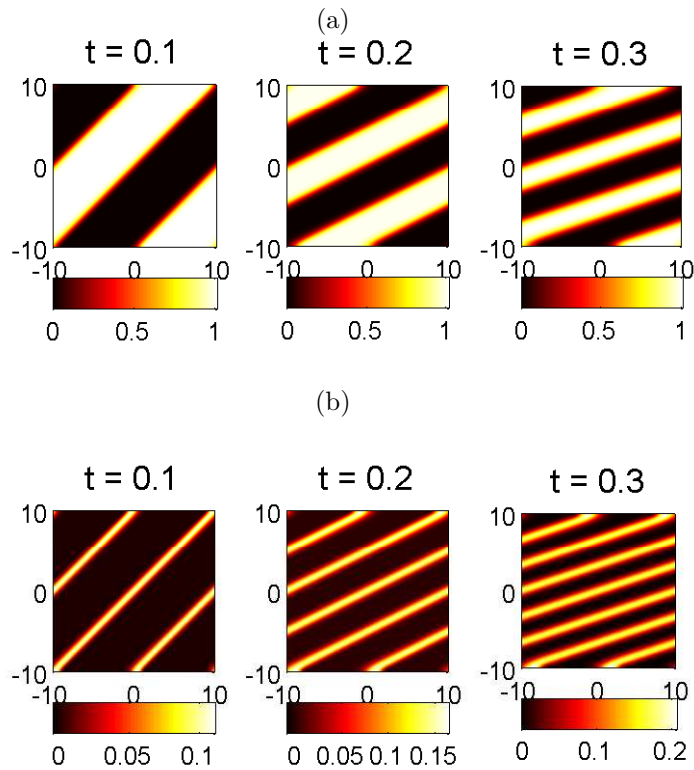


Figure B.12: (a) Surface plot of the reactant concentrations at different times during the 2D COMSOL simulation. We have chosen $Pe = 10$ and $Da = 10$ in this case. (b) Surface plot of the product concentration at different times during the 2D COMSOL simulation. We have chosen $Pe = 10$ and $Da = 10$ in this case.

lines) are in excellent agreement with each other for both the cases of $Da = 10, Pe = 1$ and $Da = 0.1, Pe = 10$.

In figure B.12 (a) and (b) we depict how the reactants and the products are stretched along the direction of the shear, leading to a corresponding compression in the perpendicular direction for times $t = 0.1, 0.2, 0.3$ for the case where $Pe = 10$ and $Da = 10$. As time progresses, the fronts from the other periodic cells start entering our cell of interest. Here at very large times, the fronts may come very close together so that the reactive mixing scales may overlap [65]. The product is formed at the interface between the two fronts and thus a larger number of bands can be observed for the surface plot of the product (figure B.12(b)).

In figure B.13, we depict the concentration distributions obtained from the COMSOL simulations in the direction normal to the interface, and compare them with those obtained from the 1D Chebyshev spectral collocation simulation. It is observed that for the case of small Péclet the number of peaks at $t = 0.35$ in the domain depicted in figure B.13 is only one while at a higher Péclet, we observe multiple peaks. It may be noted that so long as the concentration between the two peaks reaches zero, which implies independence of the two peaks, the results from the 1D simulation are valid. It may also be confirmed from figure B.13 that the concentration profiles predicted from the 1D Chebyshev spectral collocation method and full 2D simulation are in excellent agreement with each other. In the case of the 1D simulations, the boundary

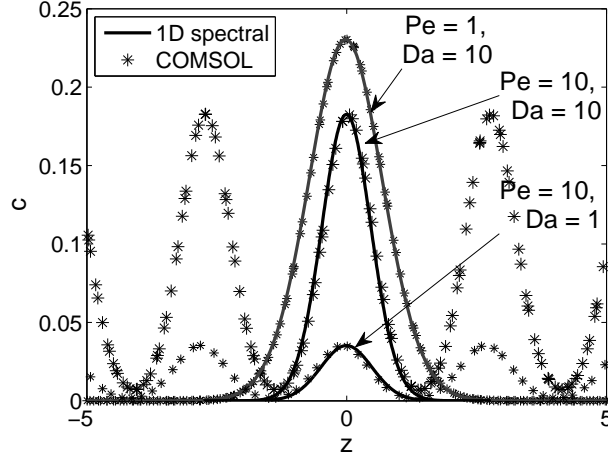


Figure B.13: Concentration profiles at $t = 0.35$, obtained in the direction perpendicular to the stretching interface from COMSOL 2D simulations and the 1D Chebyshev spectral collocation method. We depict the concentration profiles for 3 different cases $Pe = 1, Da = 10$, $Pe = 10, Da = 10$ and $Pe = 10, Da = 1$. The presence of multiple peaks in the figure from the 2D simulation is due to the fact that the periodicity employed for the 2D simulation causes the neighbouring fronts to enter the region of interest (this can be confirmed from the surface plots depicted in figure B.12).

731 condition that is implemented is in effect that of an infinite span of the domain in either directions. In the
 732 case of 2D simulations, we have taken into account the finite length of the reaction fronts. Given that the
 733 predictions of the methodology is applicable to a time before which the periodic fronts coalesce, the scenario
 734 of the two simulations are essentially the same. Hence the methodology elucidated in section 5 may be
 735 extended for complicated forms of the reactions with relative ease and with a high degree of accuracy.

736 Appendix C. Derivation of the dilution index for a conservative scalar

737 The dilution index is defined as

$$E(t) = \exp(S(t)) \quad (\text{C.1})$$

738 where $S(t) = -\int d\mathbf{x} p(\mathbf{x}, t) \ln p(\mathbf{x}, t)$ represents the system entropy and $p(\mathbf{x}, t)$ is the normalized concen-
 739 tration defined as $p(\mathbf{x}, t) = c(\mathbf{x}, t) / \int dx c(\mathbf{x}, t)$ [79]. For a conservative scalar, the analytical solution for
 740 the concentration distribution is obtained as [54]

$$c = \frac{1}{(1 + 4\theta)} \exp\left(\frac{-x'^2}{\delta^2(1 + 4\theta)}\right). \quad (\text{C.2})$$

741 Utilizing this solution, we obtain the normalized concentration and system entropy in the forms

$$p(\mathbf{x}, t) = \frac{e^{-\frac{x'^2}{\delta^2(1+4\theta)}}}{\sqrt{\pi}\sqrt{1+4\theta}\delta} \quad (\text{C.3})$$

and $S(t) = \frac{1}{2} + \frac{1}{2}\ln(1 + 4\theta) + \ln(\delta) + \frac{1}{2}\ln(\pi)$,

742 respectively, where it is noted that the form of the stretching and warped time are as of now arbitrary.
 743 Utilizing the specific results for the linear stretching, we obtain

$$E = \exp \left[\frac{1}{2} + \frac{1}{2} \ln \left(1 + 4t + \frac{4}{3} t^3 \text{Pe}^2 \right) - \frac{1}{2} \ln (1 + \text{Pe}^2 t^2) + \frac{1}{2} \ln (\pi) \right]. \quad (\text{C.4})$$

744 In order to assess the behaviour of the derivative, we attempt to find the extremum (it is observed that the
 745 extremum is a minimum). Thus, $dE/dt = 0$ yields

$$\left(\frac{1}{2} \frac{4 + 4\text{Pe}^2 t^2}{1 + 4t + \frac{4}{3} t^3 \text{Pe}^2} - \frac{\text{Pe}^2 t}{1 + \text{Pe}^2 t^2} \right) e^{\frac{1}{2} + \frac{1}{2} \ln(1 + 4t + \frac{4}{3} t^3 \text{Pe}^2) - \frac{1}{2} \ln(1 + \text{Pe}^2 t^2) + \frac{1}{2} \ln(\pi)} = 0 \quad (\text{C.5})$$

746 which implies that $\left(\frac{1}{2} \frac{4 + 4\text{Pe}^2 t^2}{1 + 4t + \frac{4}{3} t^3 \text{Pe}^2} - \frac{\text{Pe}^2 t}{1 + \text{Pe}^2 t^2} \right) = 0$. In the limit of $\text{Pet} \ll 1$, the aforementioned expression
 747 yields the corresponding time at which the minimum in dilution occurs to be, as $t = 1 / \left(2^{1/3} \text{Pe}^{2/3} \right)$, which
 748 is the same mixing time as that obtained in section 4.

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