CHEMICALLY-DRIVEN HYDRODYNAMIC INSTABILITIES

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Scientific questions

Chemical reactions can induce changes in viscosity or density which, in turn, can lead to hydrodynamic flows. Hence questions raised are:

• In which conditions can chemical reactions be at the very origin of hydrodynamic instabilities and not merely passive scalars?
• What are the properties of the patterns that can then arise?
• Influence on transport and yield of reaction?
Outline of the talk

1. $A + B \rightarrow C$ reaction fronts
2. Chemically-driven viscous fingering
3. Buoyancy-driven flows around reaction fronts
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1. $A+B\rightarrow C$ reaction fronts
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1) Reaction-diffusion $A + B \rightarrow C$ fronts

\[ \frac{\partial a}{\partial t} = D_a \nabla^2 a - kab \]
\[ \frac{\partial b}{\partial t} = D_b \nabla^2 b - kab \]
\[ \frac{\partial c}{\partial t} = D_c \nabla^2 c + kab \]

$r = B_o / A_o$

position of reaction front

$x_f = \text{position of maximum production rate } kab$
Reaction-diffusion profiles of an A+B->C front

*Galfr-Racz predictions (Phys. Rev. A 38, 3151 (1988))*

**Case 1: immobile RD front**

\[ D_a = D_b \]
\[ r = B_o / A_o = 1 \]

Position of the reaction front \( x_f = 0 \)
Case 2: moving RD front

\[ D_a \neq D_b \]

or \( r = B_o / A_o \neq 1 \)

Galfi-Racz scalings: the front moves with

\[ x_f \sim t^{1/2} \]

\[ A_o > B_o: \text{ front moves to the right (i.e. invades B)} \]
Outline of the talk

1. A+B\rightarrow C reaction fronts
2. Chemically-driven viscous fingering
3. Buoyancy-driven flows around reaction fronts
Viscous fingering instability

- The viscous fingering instability is a hydrodynamical instability that occurs when a low viscosity fluid displaces a more viscous fluid in a porous medium.

- The interface between the two fluids is unstable and develops “fingers” of the more mobile solution that invade the less mobile one.

Fingering affects both

- immiscible solutions in which case surface tension must be taken into account
- miscible solutions for which dispersion plays a key role

Here we focus on miscible solutions
Injection of a reactant A into another reactant B of the same viscosity. At the interface, production of a more viscous product C by a simple reaction A+B \rightarrow C

Rectilinear displacement in a Hele-Shaw cell (R. Maes, ULB)

CTAB injected into NaSal

B injected into A

NaSal injected into CTAB

A injected into B
Rectilinear displacement

CTAB + NaSal (dyed - initially in the cell)
$C_{\text{CTAB}} = 50 \text{ mM} - C_{\text{NaSal}} = 50 \text{ mM} - C_{\text{dye}} = 10 \text{ g/l} - U = 2.5 \text{ mm/s}$

NaSal (dyed) + CTAB (initially in the cell)
$C_{\text{CTAB}} = 50 \text{ mM} - C_{\text{NaSal}} = 50 \text{ mM} - C_{\text{dye}} = 10 \text{ g/l} - U = 2.5 \text{ mm/s}$
Theoretical model

Reactants of same viscosity

\[ \mu_A = \mu_B = \mu_0 \]

\[ \mu(c) = \mu_0 e^{\frac{R}{c_0}} \]

\[ R = \ln \left[ \frac{\mu(c = c_0)}{\mu_0} \right] \]

Symmetric case
\[ A_0 = B_0 \]
\[ D_A = D_B \]
Asymmetric case
\( B_0 = 2 \ A_0 \)
\( D_A = D_B \)

A injected into B: larger c gradient hence larger viscosity gradient

Fingering more intense where the less concentrated solution invades the more concentrated one  \( (A \ into \ B \ here) \)
B injected into A: larger c gradient hence larger viscosity gradient

Asymmetric case
\[ B_0 = A_0 \]
\[ D_A > D_B \]

Fingering more intense where the slowest species invades the quickest one (B into A here)
Rectilinear displacement

Here $D_A > D_B$

B injected into A: more intense fingering
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Buoyancy-driven flows around A+B→C fronts

Experiments in vertical Hele-Shaw cells

Density varies with concentrations

(with C. Almarcha, P. Trevelyan and P. Grosfils, ULB)
Experiments in vertical Hele-Shaw cells

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Acid-base neutralization reaction

\[ \text{HCl} + \text{NaOH} \rightarrow \text{NaCl} + \text{H}_2\text{O} \]

Visualization by interferometry
No Rayleigh-Taylor instability as acid is lighter than base here
Theoretical model

\begin{align*}
\nabla p &= -u - \rho(a, b, c)i_x,
\rho &= a + R_b b + R_c c,
\a_t + u \cdot \nabla a &= \nabla^2 a - D \, ab,
\b_t + u \cdot \nabla b &= \delta_b \nabla^2 b - D \, ab,
\c_t + u \cdot \nabla c &= \delta_c \nabla^2 c + D \, ab.
\end{align*}

\[ R_{b,c} \text{: Rayleigh numbers} \quad D = t_{hyd} / t_{chem} \]
Source of the instability:
the fast diffusion of acid towards the lower layer creates a depletion zone where locally one has “heavy over light” \(\rightarrow\) flows due to a diffusive-layer convection mechanism.
The chemical reaction breaks the symmetry of the hydrodynamic instability patterns
Active role of the color indicator
(with A. D’Onofrio et al., Buenos Aires)

Same experiment in presence of a color indicator: bromocresol green

Instability also in the lower layer due to an active role of the color indicator

Conclusions

• Chemical reactions can induce viscosity or density changes across miscible interfaces thereby influencing or creating hydrodynamic instabilities.

• The properties of hydrodynamic motions in reactive systems are different from those in non-reactive fluids.

• Very easy to add chemistry in models of hydrodynamic flows with a great number of new dynamics to uncover
1) Viscous fingering

The viscous fingering instability is a hydrodynamic instability that occurs when a low viscosity fluid displaces a more viscous fluid in a porous medium.