Development of Concentration-Dependent Diffusion Instability in Reactive Miscible Fluids under Influence of Constant or Variable Inertia

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In last decades the interaction between reaction-diffusion phenomena and pure hydrodynamic instabilities has attracted increasing interests because the chemically-induced changes of fluid properties such as density, viscosity or surface tension may result in the instabilities, which exhibit a large variety of convective patterns.

Scenario for instability development essentially differs for immiscible and miscible fluids. The simple, irreversible chemical scheme such as a neutralization reaction A+B->S occurring in binary liquid-liquid immiscible systems was studied in [1-4]. The irregular plumes and fingers commonly observed here because of more dense acid on top of less dense base is unstable under gravity via the Rayleigh-Taylor (RT) mechanism [2]. A much more impressive, regular pattern of cellular-like fingers arising near the interface was reported for an organic base [1]. The regularity was shown to originate from the perfect balance between the RT instability and the Rayleigh-Bénard [3] and Marangoni mechanisms [4]. Thus, a liquid-liquid interface was recognized to be important for fine-tuning of salt fingers.

A completely different situation was observed in the miscible case. The main engine breaking the equilibrium here was found to be the difference between the diffusion rates of species resulting in double diffusive instability or diffusive-layer convection as well as RT instability [5]. The works dealing with this subject, have usually reported the formation of irregular patterns of fingers.

Absolutely in all the works in this field to our knowledge the diffusion coefficients of species have been assumed to be constant. Generally, a concentration-dependence exists in most systems, but often, e.g. in dilute solutions, the dependence is slight and the diffusion coefficient can be assumed constant for practical purposes.

In this work, we focus on the processes which accompany a frontal neutralization reaction occuring between two miscible fluids. We report a new type of instability, *the concentration-dependent diffusion* (CDD) instability that arises when the diffusion coefficients of species depend on their concentrations.

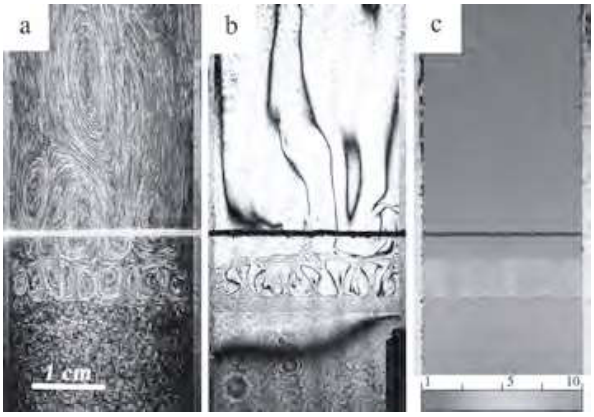

Figure 1. Chemo-convective structures arising due to the CDD instability observed 1100 s after as the aqueous solutions of HNO_3 (top) and NaOH (bottom) were brought into contact in a Hele-Shaw cell under constant gravity: (a) Velocity field; (b) Interferogram; (c) *p*H distribution. Initial concentrations are equal to 1 mol/l.

We demonstrate both experimentally and theoretically that chemically-induced changes of reagent concentrations coupled with concentration- dependent diffusion can produce spatially localized zone with unstable density stratification that under the gravity gives rise to the development of perfectly periodic convective structure even in a miscible system (Fig.1). We have tested a number of other systems and have found a similar patterning there. In our opinion, it may indicate that the discovered effect is of a general nature and should be taken into account in reaction-diffusion-convection problems as another tool with which the reaction can govern the movement of the reacting fluids.

Since this new type of instability is sensitive to inertial field, we investigate the influence of periodic vibrations on the non-linear development of instability both in the case of low and high-frequency vibrations.

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References
[1] K. Eckert, M. Acker, Y. Shi., *Phys. Fluids*, 16 (2004) 385.
[2] D.A. Bratsun, A. De Wit, *Chem. Eng. Sci.*, 66 (2011) 5723.
[3] D.A. Bratsun, A. De Wit, *Phys. Fluids*, 16 (2004) 1082.
[4] D.A. Bratsun, *Microgravity Sci. Technol.*, 26 (2014) 12.
[5] P. M. J. Trevelyan, C. Almarcha, A. De Wit, *Phys. Rev. E* 91 (2015) 023001.