Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Marangoni- vs buoyancy-driven flows : competition for spatio-temporal oscillations in A + B → C systems

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Movies SM1.a-b display the complete spatio-temporal evolution of the concentration field of the product C for the systems represented in Fig. 1a and Fig. 1b respectively, and described in section 3.1.

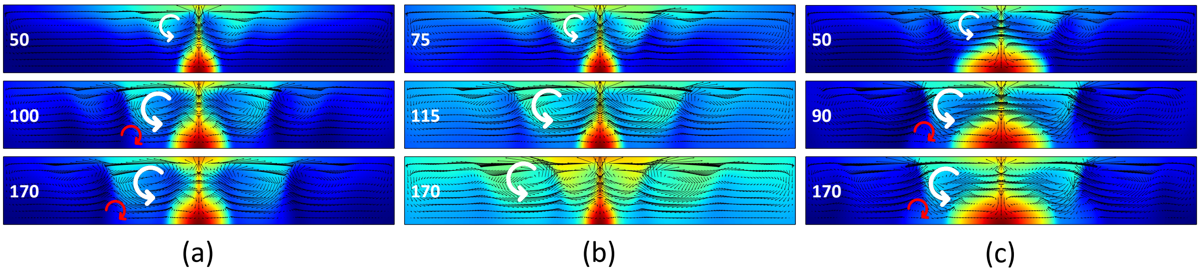
Movies SM2.a-b show the spatio-temporal evolution of the concentration field of the product C for non-oscillating systems when one of the parameters ΔM or ΔR is predominant. SM2.a ($\Delta M = -400, \Delta R = -0.25$) represents a system where the Marangoni-driven convection is predominant, while SM2.b ($\Delta M = -100, \Delta R = -2.25$) represents the opposite case where the buoyancy-driven convection is predominant.

Movies SM3.a-b display the complete spatio-temporal evolution of the vertical velocity field for the unimodally (a) and bimodally (b) oscillating systems represented in Fig. 7 left and right respectively. **Movies SM3.c-d** show the corresponding spatio-temporal evolution of the product concentration field for the unimodally (c) and bimodally (d) oscillating systems.

Fig. SF1 gives complementary information regarding the apparition of convective rolls. The figure displays concentration field of the product C for three systems at specific times and illustrates the interaction of the fingers with the product located at the bottom. In SF1.a ($\Delta M = -300, \Delta R = -2$), the system evolves over time with the emergence of fingers created at the surface and reaching the bottom of the reactor. The product C located at the reactor's bottom due to the sinking is localised, and weak interactions with the fingers generate new convective rolls of small amplitude at the bottom of the reactor. However, no bimodal oscillations are observed in this case as the extent of the buoyancy-driven convective rolls does not allow for a further amplification of the newly formed convective rolls by interacting with them.

As $|\Delta M|$ increases (see SF1.b for $\Delta M = -500, \Delta R = -2$), the extent of the fingers does not change, but oscillation's frequency increases. The product C is here also localized in a narrow region at the center of the system, preventing any interaction.

In contrast, if $|\Delta R|$ increases (SF1.c for $\Delta M = -500, \Delta R = -5$) the product located at the center of the system is slowly spreading out at the bottom boundary, enabling its interaction with the fingers. This interaction is followed by the emergence of new counter-rotating convective rolls, leading to bimodal oscillations, as described in section 3.2.2.



SF 1 – Snapshots of the concentration field of the product C at specific times for : (a) $\Delta M = -300$, $\Delta R = -2$; (b) $\Delta M = -500$, $\Delta R = -2$ and (c) $\Delta M = -500$, $\Delta R = -5$. The white arrows represent the Marangoni-driven convective rolls and the red arrows represent the newly merged convective rolls.