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A TRANSFER OPERATOR BASED COMPUTATIONAL STUDY OF REACTING FLUIDS

We analyze transport and reactions of ideal fluid particles. We assume that they move passively according to $\dot{x}(t) = u(x(t), t)$, with tracer trajectories $x(t) \in \mathbb{R}^2$. Let $S: M \to M$ be the flow map that maps a particle $x_0 = x(t_0)$ to its new position $S(x_0) = x(t_0 + \tau)$ over the time span $[t_0, t_0 + \tau]$.

In practice, we represent fluids by density vectors, which are evolved by means of a numerical transfer operator and are subsequently updated according to an underlying chemical reaction scheme. We model a stirred tank reactor (STR) as a compact domain $M \subset \mathbb{R}^2$ discretized into n disjoint, connected sets (boxes) B_i, $i = 1, \dots, n$ [1]. Each box B_i contains N uniformly distributed sample points of chemical substances $x^i_{0,k}, k = 1, \dots, N$. We use Ulam's method [2] for a finite-rank approximation of an infinite-dimensional Perron-Frobenius operator as a matrix $\bar{P} \in \mathbb{R}^{n,n}$ as follows:

$$\bar{P}_{ij} = \frac{\#\{k:\ S(x^i_{0,k}) \in B_j\}}{N},$$

i.e. the entry $\bar{P}_{ij}$ is estimated as the proportion of the fluid particles that gets mapped from box B_i to box B_j under the action of S. We can interpret the row-stochastic matrix $\bar{P}$ as the transition matrix of a Markov chain on a finite state space, where the boxes B_i represent the states. A fluid density vector $C_{A_t} \in \mathbb{R}^n$ representing a chemical substance A at time t evolves according to $C_{A_{t+\tau}} = C_{A_t}\bar{P}$. In nonautonomous systems $\bar{P}$ depends explicitly on time.

Double Gyre Mixer (DG)

The DG mixes the fluids over the domain [0,2]×[0,1] with the velocity field u [3] defined by

$$\dot{x} = -0.5\pi \sin(\pi f(x,t)) \cos(\pi y)$$

$$\dot{y} = 0.5\pi \cos(\pi f(x,t)) \sin(\pi y) \frac{df}{dx}(x,t),$$

where $f(x,t) = \epsilon \sin(2\pi t) x^2 + (1 - 2\epsilon \sin(2\pi t)) x$. We differentiate between an autonomous ($\epsilon = 0$) and a time dependent system with $\epsilon = 0.4$.

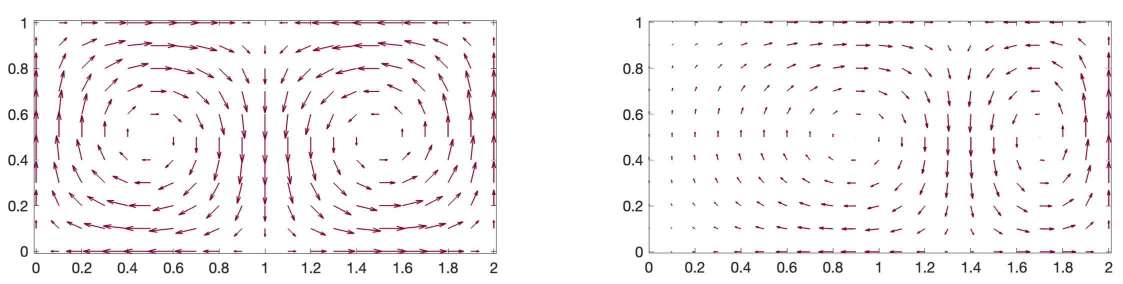

Fig. 1: Flow velocity field in an autonomous system (left) and in a time dependent system at $t = 0.25$ (right).

Van de Vusse's Competitive Consecutive Reaction [4]

$$A \overset{k_1}{\to} B \overset{k_2}{\to} C$$

$$2A \overset{k_3}{\to} D$$

The constant feed concentration C_{A0} of pure A is continuously fed into the STR (volume V per box, flow rate v_0) and the concentrations C_i are pumped out of the STR (red: input/output dynamics, see fig. 2). In each box B_i, the particles react under the following conditions:

$$\dot{C_A} = (C_{A0} - C_A) \cdot \frac{v_0}{V} - k_1 C_A - k_3 C_A^2$$

$$\dot{C_B} = -C_B \cdot \frac{v_0}{V} + k_1 C_A - k_2 C_B$$

$$\dot{C_C} = -C_C \cdot \frac{v_0}{V} + k_2 C_B$$

$$\dot{C_D} = -C_D \cdot \frac{v_0}{V} + \frac{1}{2} k_3 C_A^2,$$

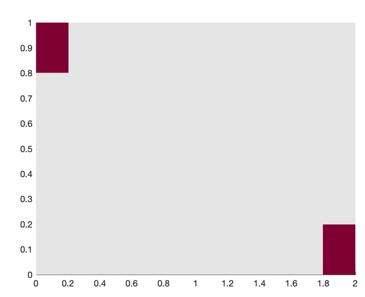

Fig. 2: Input area (northwest) and output area (southeast).

where $\pm C_i$ red is the input/output on the input/output area.

Competitive Consecutive Reaction in an Open DG Reactor Model

We consider a STR with $V = 1l$, $C_{A0} = 10\,mol/l$ and $v_0 = 10\,l/min$. For each time step $\tau = 0.01$, the particles are transported and then react (here $k_1 = \frac{5}{6}$, $k_2 = \frac{5}{3}$ and $k_3 = \frac{1}{6}$). The reaction is simulated using the explicit Euler method.

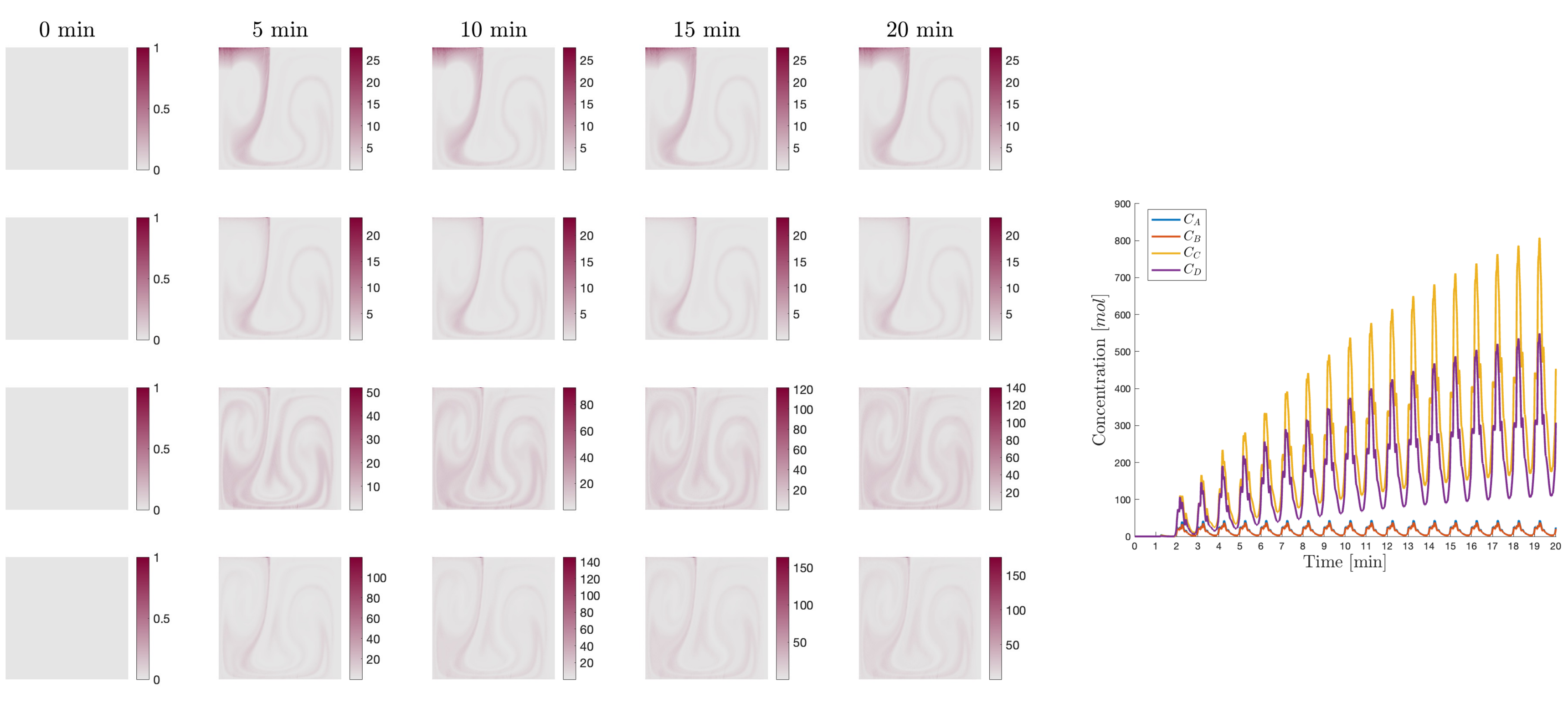

Fig. 3: Competitive consecutive reaction with $C_A, C_B, C_C, C_D [mol]$ (row 1 to 4, left) and output concentrations (right) in an open time dependent system.

Competitive Reaction

$$A + B \to C$$

$$B + C \to W$$

After each unit time step, in each box $\mathrm{B_i}$ the particles react under the following conditions:

If $C_{B_t} \geq C_{B_{t-1}} - min\{C_{A_{t-1}}, C_{B_{t-1}}\} - min\{C_{B_{t-1}}, C_{C_{t-1}}\}$:

$$C_{A_t} = C_{A_{t-1}} - min\{C_{A_{t-1}}, C_{B_{t-1}}\}$$

$$C_{B_t} = C_{B_{t-1}} - min\{C_{A_{t-1}}, C_{B_{t-1}}\} - min\{C_{B_{t-1}}, C_{C_{t-1}}\}$$

$$C_{C_t} = C_{C_{t-1}} + 2\,min\{C_{A_{t-1}}, C_{B_{t-1}}\} - min\{C_{B_{t-1}}, C_{C_{t-1}}\}$$

$$C_{W_t} = C_{W_{t-1}} + 2\,min\{C_{B_{t-1}}, C_{C_{t-1}}\},$$

i.e. particles B react to both C and W.

If $C_{B_t} < C_{B_{t-1}} - min\{C_{A_{t-1}}, C_{B_{t-1}}\} - min\{C_{B_{t-1}}, C_{C_{t-1}}\}$ the particles B in box B_i react either with A to C or with C to W.

Competitive Reaction in a Closed DG Reactor Model

We consider a STR that is filled with reactants A and B in selected boxes at time t_0 with concentrations $C_i = 1\,mol$ per box. For each time step $\tau = 1$, the particles are first transported and then react.

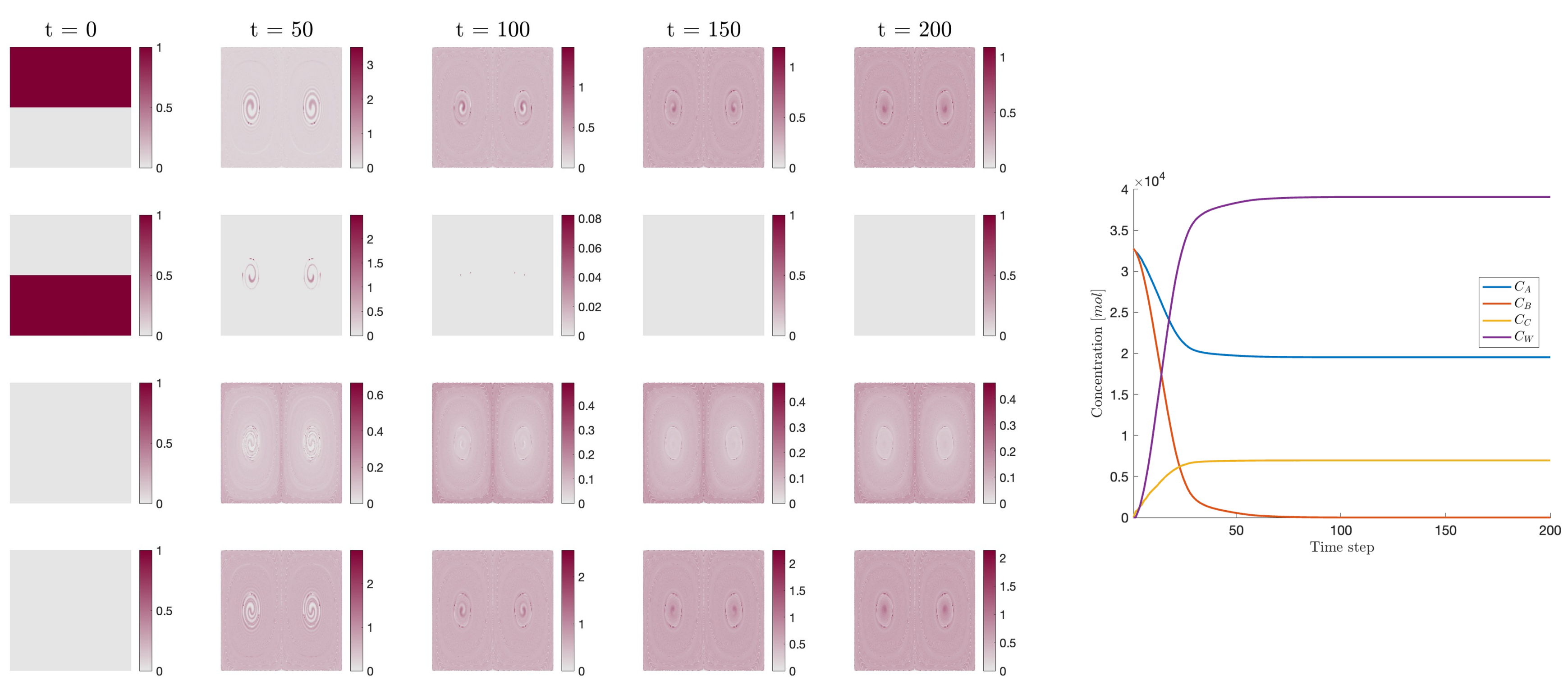

Fig. 4: Competitive reaction with $C_A, C_B, C_C, C_W [mol]$ (row 1 to 4, left) and its concentrations (right) in a closed autonomous system.

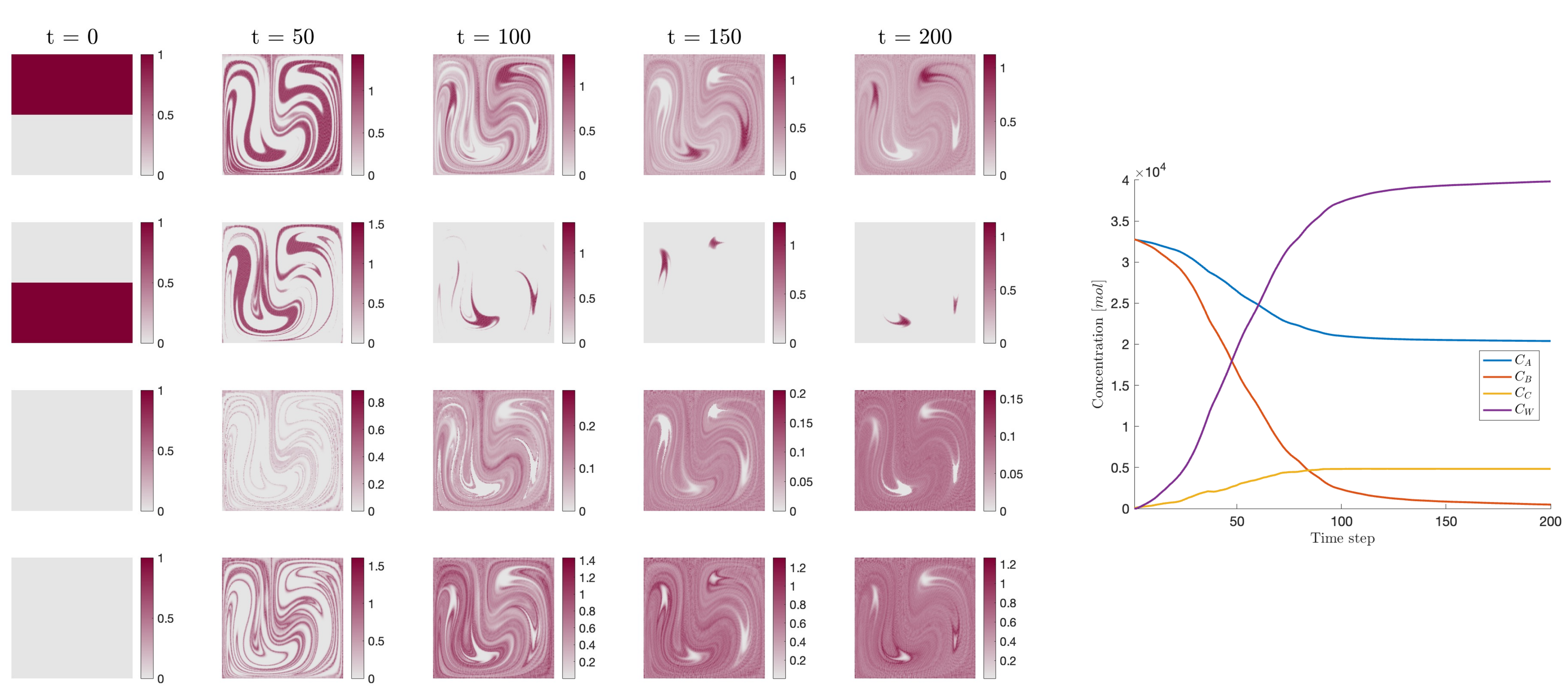

Fig. 5: Competitive reaction with $C_A, C_B, C_C, C_W [mol]$ (row 1 to 4, left) and its concentrations (right) in a closed time dependent system.

Outlook

In the future, we will implement a diffusion into neighboring boxes and compare experimental data with the transfer operator based results and visualize the distribution of particles and their concentrations as networks.

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