Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information for

Control over the Formation of Supramolecular Material Objects Using Reaction-Diffusion

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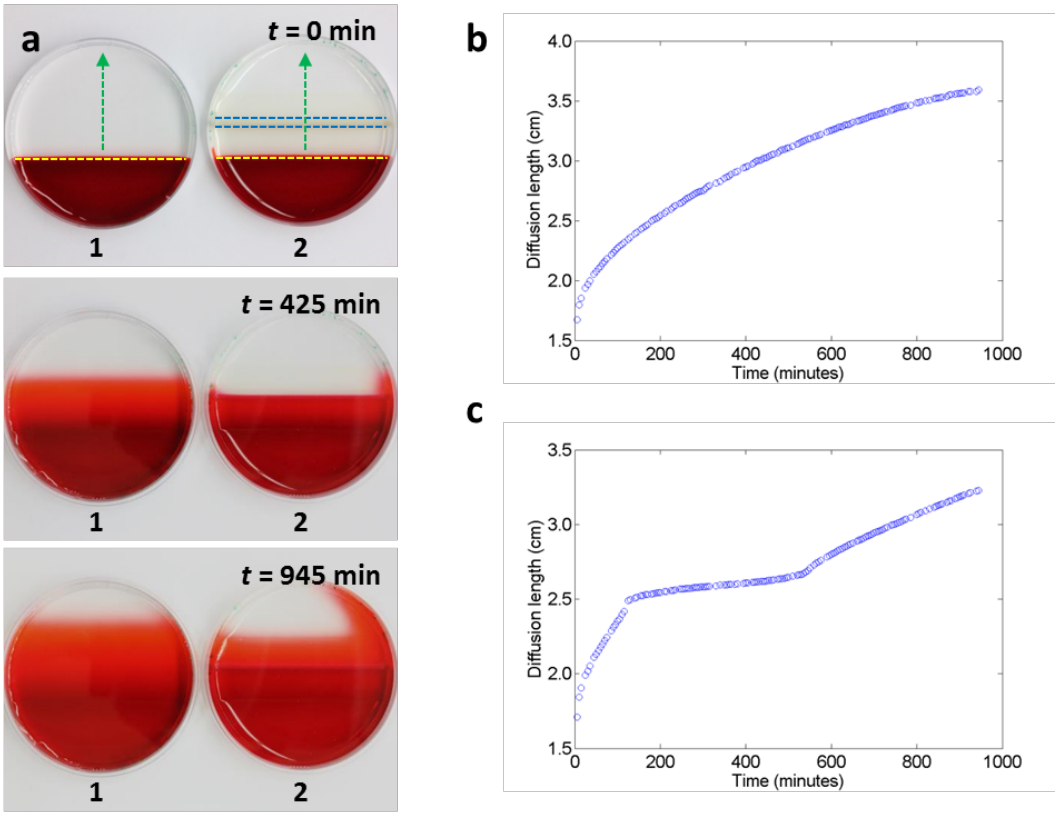
Slow down of diffusion by the formation of the supramolecular pattern and its implementation in the RD model

Initial simulations, in which we ignored dynamic variations in the diffusion coefficients, showed significant discrepancies between the experimental data and the model at later stages of pattern development. Whereas in experiments w_{HA3} reached a constant value after initial growth, the model showed a continuous increase of w_{HA3}. Based on this observation we anticipated that after initial formation, the **HA_3** structure hampers further diffusion of **H** and **A**. To test this hypothesis, we let a food-grade dye (chemical composition unknown) diffuse through a blank agar matrix and an agar matrix with a preformed supramolecular structure inside. We observed that the diffusion of the dye was slower in the region with the supramolecular structure, see Supplementary Figure 1.



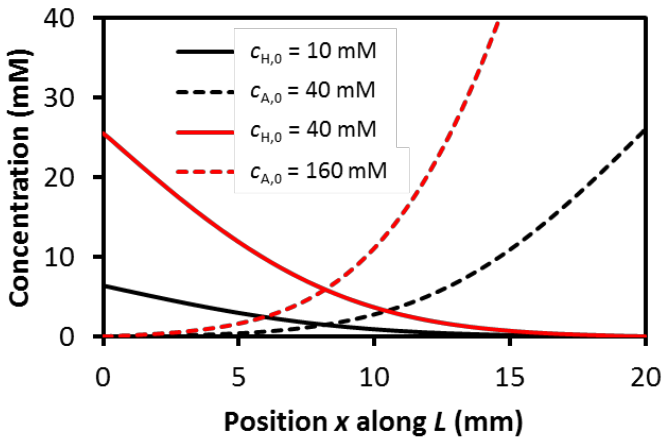
Supplementary Figure 1. Effect of a supramolecular pattern on the diffusion of a food-grade dye (chemical composition unknown) through 1% agar. a) Photographs of the diffusion of the food-grade dye through 1% agar without and with a supramoleculer pattern at different time points. The Petri dish **1** contained the agar matrix without supramolecular patter, whereas the Petri dish **2** contained the agar matrix with the supramolecular pattern. The interface between the reservoir containig the dye and the agar is marked with the yellow dashed line. The edges of the supramolecular pattern are marked with the blue dashed lines. Diffusion length was determined along the distance indicated with the green arrow. The Petri dishes were 5 cm in diameter. **b)** Distance traveled in 1% agar. **c)** Distance diffused in 1% agar with the supramolecular patter. The food-grade dye used was purchased in a supermarket and diluted 4 times. It was determined from the graphs **b** and **c** that the diffusion coefficent of the food-grade dye in the 1% agar and the supramolecular pattern is 3.6×10^{-9} m^2 s^{-1}, and 4.5×10^{-10} m^2 s^{-1}, respectively.

Based on this finding, we introduced a space-time dependency of $D_{\mathbf{H}}$, $D_{\mathbf{A}}$, $D_{\mathbf{HA}}$ and $D_{\mathbf{HA2}}$ based on the spatio-temporal concentration of **HA_3**. The decrease was defined using the stretched exponential function

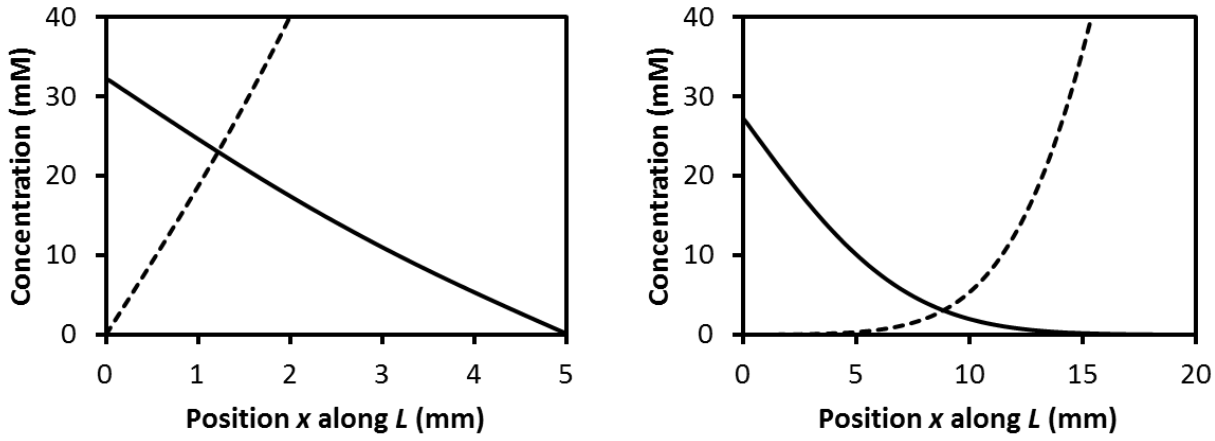
$$D_g = D_0 \exp\left(-a\varphi^{\upsilon}\right)$$

with D_g and D_0 being the diffusion coefficients of the species in gel and in water solution (diffusion in 1% agar is negligibly different from diffusion in pure water), respectively, and φ is the volume fraction of **HA_3**. Parameter υ was set to 1 for simplicity and parameter a was determined by fitting the model to the experimental data. The parameter a was set to 30 for **H**, and 34 for **A**, **HA**, and **HA_2**. With the slow down of diffusion due to the formation of the supramolecular pattern implemented, we were able to reproduce the experimentally observed behaviour using the model.

Concentration gradients



Supplementary Figure 2. Effect of initial concentration of H and A on their respective concentration for L = 20 mm after 12 hours of diffusion, where first pattern formation was observed. The initial concentraton of H and A were denoted as $c_{H,0}$ and $c_{A,0}$. This graph was generated by solving equations 1 and 2 without the reaction term, *i.e.* all reaction constants were 0, to illustrate concentration profiles purely based on diffusion.



Supplementary Figure 3. Effect of diffusion distance on concentration profiles of H and A for *L* = 5 mm and *L* = 20 mm at the moment of partial structure formation (3h for *L* = 5 mm and 8h for *L* = 20 mm). The initial concentratons of **H** and **A** were denoted as $c_{H,0}$ and $c_{A,0}$. This graph was generated by solving equations 1 and 2 without the reaction term, *i.e*. all reaction constants were 0.