Transition of Liesegang Precipitation Systems: Simulations with an Adaptive Grid PDE Method

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Abstract. The dynamics of the Liesegang type pattern formation is investigated in a centrally symmetric two-dimensional setup. According to the observations in real experiments, the qualitative change of the dynamics is exhibited for slightly different initial conditions. Two kinds of chemical mechanisms are studied; in both cases the pattern formation is described using a phase separation model including the Cahn-Hilliard equations. For the numerical simulations we make use of an adaptive grid PDE method, which successfully deals with the computationally critical cases such as steep gradients in the concentration distribution and investigation of long time behavior. The numerical simulations show a good agreement with the real experiments.

1 Introduction

It is more than 110 years that Liesegang observed and reported an interesting phenomenon [23]: the precipitate in some simple chemical reactions may not homogeneously distribute. In the typical experimental setup, one chemical reagent is uniformly distributed in a gelled medium (called inner electrolyte), while the other one (called outer

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electrolyte) diffuses from outside. The initial concentration of the outer (invading) is chosen to be much larger than that of inner one. This condition ensures the higher diffusion flux of the outer electrolyte into the gel. In some circumstances, in the wake of the chemical front some precipitation bands are formed, following each other. In 1D the distances between the bands are determined by the geometrical law [19], see Fig. 1. For the description of this phenomenon many models have been proposed such as models based on simple supersaturation [20] or competitive particle growth [8, 15] and models based on phase separation [2, 3, 27]. A general framework for the different models in 1D has been recently published [30]

As the pattern formation in 2D has recently gained a great interest in the engineering of microsystems [17], a number of experimental studies have been performed. Interestingly, different dynamics have been reported for similar-centrally symmetric-experimental setups: in many cases a regular Liesegang pattern evolved [21, 22], in other experiments only one moving precipitation layer was detected [28, 31].

Our aim is to exhibit and reproduce this phenomenon with numerical simulations and to point out that this can happen using the same material coefficients with a slight modification of initial conditions.

For a successful simulation procedure we have to choose

- an adequate model of the underlying chemical mechanism;

- an effective numerical method for solving the PDE for the evolution.

Among the possibilities mentioned above, we have chosen the phase separation model proposed in [2], where the time evolution of the precipitate is described with the Cahn-Hilliard equations, which was originally proposed in [7].

According to this model, the precipitate segregates into the low and high density phases if its local concentration reaches a critical threshold ("spinodal point"). The corresponding fourth order PDE serves as an accurate model: the empirical laws for the Liesegang patterns have been verified by numerical simulations [2] in a one dimensional setup. The dynamics driven by the Cahn-Hilliard equations have been analyzed in a series of studies, see, e.g., [1, 4, 5] and is still in the focus of theoretical investigations. Note that we investigate the Cahn-Hilliard equation within a reaction-diffusion system.

In the real applications, the regions which are used as a source of one of the reactants are small compared to the scale of the computational domain. This results in difficulties in the traditional numerical simulations due to the high concentration gradient of the outer (invading) electrolyte and precipitate. On the other hand, frequently, pattern formation phenomena have to be simulated over a relatively long time period. In this way, an overly accurate space discretization or too short time steps can easily result in very time consuming simulations. Therefore, it is important to apply an accurate and fast numerical solver, which successfully deals with the above difficulties. Several techniques can improve the computational procedures such as (i) using appropriate numerical integrators; (ii) using parallel program environment (supercomputer, cluster, GRID systems [24] or video card using specially designed program environment (CUDA) [29]),