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Numerical approximation of Turing patterns in electrodeposition by ADI methods

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ABSTRACT

In this paper we study the numerical approximation of Turing patterns corresponding to steady state solutions of a PDE system of reaction–diffusion equations modeling an electrodeposition process. We apply the Method of Lines (MOL) and describe the semi-discretization by high order finite differences in space given by the Extended Central Difference Formulas (ECDFs) that approximate Neumann boundary conditions (BCs) with the same accuracy. We introduce a test equation to describe the interplay between the diffusion and the reaction time scales. We present a stability analysis of a selection of time-integrators (IMEX 2-SBDF method, Crank–Nicolson (CN), Alternating Direction Implicit (ADI) method) for the test equation as well as for the Schnakenberg model, prototype of nonlinear reaction–diffusion systems with Turing patterns. Eventually, we apply the ADI-ECDF schemes to solve the electrodeposition model until the stationary patterns (*spots & worms* and *only spots*) are reached. We validate the model by comparison with experiments on Cu film growth by electrodeposition.

1. Introduction

[...] *Is the discrete world an approximation of the continuous one or is it the other way around?* This sentence echoes another famous one, stated 40 years ago by Eugene Wigner: *The miracle of appropriateness of the language of mathematics for the formulation of laws of physics is a wonderful gift which we neither understand nor deserve* [1]. Discrete and continuous descriptions of a given phenomenon can be, rather crudely, regarded as the expression of the same concept in two different languages. Nevertheless, there is something unique in both approaches. At least from the mathematical point of view, the uniqueness of continuous and discrete worlds cannot be entirely captured by the straightforward concepts of analytical and numerical treatments of equations, respectively. Probably, such uniqueness resides in the modeling choices that are an aspect of the cognitive reduction of a phenomenon that seems to be unavoidable in order to set up a quantitative treatment. Of course, this view of discretization has a bearing on both model building and numerical approximations. In fact, in the contemporary language of mathematics, a special place is occupied by numerical analysis, thanks to the steadily developing interest for the interplay among abstract formalism, computations and simulation of real world phenomena.

This contribution means to offer an example of such conceptual interplay made possible by the synergy of advanced materials-science problems with clear-cut technological relevance, well-defined mathematical formulation of the underlying physics and suitable computational methods. Specifically, the problem we attack in this study is part of a long-term project dealing with the continuous mathematical modeling of metal growth by electrodeposition (ECD), whose

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previous results are reported in [2–10]. In these studies, we have introduced a reaction–diffusion PDE system, accounting for the coupling between morphology and surface concentration of one chemical species adsorbed at the surface of the growing metal. Morphology and surface chemistry have been considered as *continuous* variables. This system exhibits a surprisingly rich dynamic scenario, featuring: (i) existence of transition front waves moving with specific wave speeds; (ii) Turing instability and initiation of spatial patterns driven by diffusion; (iii) smoothing effects related to a forcing sinusoidal term.

In all cases, a numerical *discretization* for the electrochemical PDE system is needed to gain quantitative information on the evolution of the solution until its steady state is attained. As far as traveling wave solutions are concerned, in [7] we have proposed an accurate approximation of the wave profile and of its speed. Concerning the simulation of Turing patterns, in one [2,4] and two [5,6,8,9] space dimensions, we have used a general-purpose scheme in order to map the dynamic behavior and confirm numerically the outcomes of theoretical stability analyses.

Here we propose to extend the numerical method introduced in [11,12] and developed for the approximation of traveling waves in [7], to deal with Turing patterns. This approach consists in a high order semi-discretization in space by the Extended Central Difference Formulas (ECDFs). As far as the discretization in time is concerned, we discuss the appropriateness of selected numerical techniques: the Crank–Nicolson (CN) method, the Implicit–Explicit (IMEX) Semi-Backward Differentiation Formula of order 2 (2-SBDF) and the Peaceman–Rachford Alternating Direction Implicit (ADI) scheme. For this purpose, we introduce a linear test reaction–diffusion equation, given by a heat equation with linear source term, and define its stability region in terms of reaction and diffusion scales. Then the stability regions for the numerical methods are derived. This analysis allows to identify stepsize restrictions and to decide which method is best suited. The results on stepsize restrictions are applied to the Schnakenberg model, prototype of nonlinear reaction–diffusion systems with Turing patterns (see [13]).

Our study is completed by a comparison of the numerical simulations (discrete results) with experimental data (continuous observables). Their consistency can be regarded as a factual instance of Donato Trigiante's tenet that *the qualitative behavior of the solution of the continuous problem and the qualitative behavior of the discrete one must be similar* (see [14, Chapter 1, p.1]).

The paper is organized as follows. In Section 2 we highlight the mathematical model for metal growth by electrodeposition and the analytical results to guarantee Turing instability and pattern formation. In Section 3 we recall the ECDF schemes in 1D and their extension in 2D together with some properties about the matrix operator for the derivative approximations. In the same section, we set up the ODE system arising from the semi-discretization in space by ECDFs ready for subsequent processing by a time integration method. In Section 4 we present the test reaction–diffusion problem, the stability analysis for the time integration schemes cited above, their possible stepsize restrictions and numerical results for the Schnakenberg model. In Section 5, we show the numerical results obtained by the ADI-ECDF schemes of order $p = 2, 4$ for the approximation of the Turing patterns for the ECD model and we address also the model validation through comparison between simulations and experiments.

2. The continuous model

Metal plating is a well-assessed and widespread technology present in several fields from heritage to nuclear science and aerospace. In fact, it is ubiquitous in surface treatment technologies and exhibits a wide-range of applications including, among others: energetics (fuel cells and batteries), chemical and biochemical sensors, electronic fabrication, corrosion and wear protection, surface nobilitation and decoration, preservation of metallic components. Usually, the functional and aesthetic quality of metal coatings is achieved on empirical basis. Recently, starting from the paper [2], the present authors have proposed a tentative rationalization of the above process by introducing a system of coupled reaction–diffusion equations for the description and prediction of morphogenesis of the electro-deposits at the electrochemical interface during metal plating at controlled potential. These initial results opened the way to a series of other papers [3–8,10] where we focus on different kinds of continuous solutions giving rise to waves moving with specific speed and pattern formation. In this system one equation is for the morphology and one for the surface concentration of a key adsorbate. Hence, we describe the evolution of the electrodeposit surface profile obtained as the solution of a balance equation. The flow terms describe inflow and outflow of material contributing to the build-up of the morphology, while the source terms account for generation (deposition) and loss (corrosion, desorption) of the relevant material. In the rest of this section we review our key results, leaving out the details, that can be recovered from the original papers.

For the electrokinetic reasons detailed in [6,7], the relevant PDE system of reaction–diffusion equations with zero Neumann boundary conditions (BCs) for the electrode morphology $\eta(x, y, t)$ and the surface chemistry coverage $\theta(x, y, t)$ is given on a representative domain $\Omega = \Omega_x \times \Omega_y \subset \mathbb{R}^2$ by:

$$\begin{cases} \dfrac{\partial \eta}{\partial \tau} = D_\eta \Delta \eta + A\dfrac{\eta^2}{1+\eta} - B\eta\theta, & \tau > 0 \\ \dfrac{\partial \theta}{\partial \tau} = D_\theta \Delta \theta + (1-\theta)K_{ADS}(\eta,\theta) - \theta K_{DES}(\eta,\theta), & \\ (\mathbf{n}\cdot\nabla\eta)|_{\partial\Omega} = (\mathbf{n}\cdot\nabla\theta)|_{\partial\Omega} = 0, & \\ \eta(x,y,0) = \eta_0(x,y), \theta(x,y,0) = \theta_0(x,y), & (x,y) \in \Omega \end{cases} \tag{1}$$