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MECHANICAL EFFECTS ON TURING PATTERN FORMATION IN BULK DIFFUSION AND SURFACE REACTION-DIFFUSION SYSTEMS

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Abstract. *The coupling between bulk diffusion and surface reaction-diffusion is a critical factor underlying many complex behaviors observed in fields like material science and biology, including phenomena such as chemical pattern formation and oscillations. This study investigates how this coupling is influenced by mechanical effects resulting from bulk diffusion occurring within an elastic solid. In particular, we address the role played by this mechanochemical interplay in the formation of Turing's patterns using a reaction kinetics model of the Fitzhugh–Nagumo type. To achieve this objective, we perform numerical simulations of the nonlinear system of equations using a mixed finite element implementation in the COMSOL Multiphysics platform. The proposed numerical formulation is applied to the investigation of pattern formation on the surface of a sphere. The results suggest that a pressure load applied to the surface has a stabilizing effect, suppressing pattern formation in the bulk variable.*

Keywords: *bulk-diffusion, surface reaction-diffusion, Fitzhugh–Nagumo kinetics, mechanochemical coupling.*

1. INTRODUCTION

The interaction between bulk diffusion and surface processes, including chemical reactions and diffusion at the boundary of the bulk domain, is crucial in many scientific and technological fields. One instance is the development of solid-state hydrogen storage materials, where the slow kinetics of hydrogen uptake and release by metals are attributed to this interplay. The kinetics of hydrogen uptake from a gaseous environment involves the adsorption and dissociation of hydrogen molecules and hydrogen penetration into and diffusion within the solid (Martin *et al.*, 1996). Another complex phenomenon that relies on the interplay of bulk diffusion and surface processes is protein pattern formation in cell biology (Burkart *et al.*, 2022). In this case, the dynamics of intracellular signaling molecules such as Cdc42, which exists in inactive and active forms (Gomez *et al.*, 2021), capture essential features of the phenomenon. The inactive form diffuses through the cytoplasm and membrane, undergoing attachment, detachment, and interconversion with the active form, which can also diffuse.

In addition to the interaction between bulk and surface, through diffusion and chemical reactions, the mechanical deformation of the body also plays a fundamental role in such processes. The study of deformations induced by the presence and transport of chemical species within a solid has been the focus of many researchers, due to its potential for applications in areas such as robot engineering of active soft materials (Miller and Dunkel, 2020) and also because it is an important mechanism involved in the phenomena of cellular polarization (Mao and Lecuit, 2016). In soft active materials, traveling waves, generated by reaction-diffusion processes, can locally contract, shear, or even deform the surfaces on which they propagate (Miller *et al.*, 2018). At the cellular and tissue level, the formation of chemical and mechanical patterns is fundamental in processes such as embryogenesis, tissue formation, and the development of some diseases (Brinkmann *et al.*, 2018; Hannezo and Heisenberg, 2019; Bailles *et al.*, 2022). In the context of industrial applications, the presence of hydrogen inside the metal is responsible for reducing the material's resistance. This phenomenon is known as hydrogen embrittlement and is one of the main limitations for the safe use of hydrogen as an energy source (Dwivedi and Vishwakarma, 2018; Meda *et al.*, 2023). The process of hydrogen ingress into metals from the environment is also a phenomenon influenced by the mechanical contribution (Hageman and Martínez-Pañeda, 2022).

In this work, we investigate the influence of mechanical effects in mechanochemical problems that display a coupling between bulk diffusion and surface reaction-diffusion. In particular, we investigate the emergence of Turing's patterns on the surface of an elastic solid.

2. MECHANOCHEMICAL MODEL

In this work, we will explore the mechanochemical interplay discussed by Duda *et al.* (2023). Consider a region $\mathcal{R}$ occupied by a deformable solid body in its reference configuration. Within this body, a constituent B is able to move freely through its interstices while simultaneously inducing deformation. On the boundary of this region, $\partial\mathcal{R}$, the constituents A and C can move freely and react with each other. The surface is coupled to the bulk by the consumption/generation of the bulk constituent through adsorption/desorption. Limiting the analysis to situations involving infinitesimal strains and rotations, this particular mechanochemical coupling is described by the following set of equations:

$$\left.\begin{aligned}
&\left.\begin{aligned}
&\operatorname{div}\big(\lambda(\operatorname{div}\boldsymbol{u})\boldsymbol{I}+G(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\top})-\kappa\upsilon(n_{B}-n_{\mathrm{ref}})\boldsymbol{I}\big)=\boldsymbol{0},\\
&\dot{n}_{B}=-\operatorname{div}\Big(-D_{B}\nabla n_{B}+\frac{D_{B}\upsilon}{k_{B}T}n_{B}\nabla\sigma\Big),
\end{aligned}\right\}\quad\text{in}\quad\mathcal{R};\\
&\left.\begin{aligned}
&\big(\lambda(\operatorname{div}\boldsymbol{u})\boldsymbol{I}+G(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\top})-\kappa\upsilon(n_{B}-n_{\mathrm{ref}})\boldsymbol{I}\big)\boldsymbol{n}=\boldsymbol{s}_{\mathrm{env}}\\
&\frac{\hat{K}(\sigma)n_{B}}{\bar{n}_{B}+\hat{K}(\sigma)n_{B}}=\frac{n_{A}}{\bar{n}_{A}},\\
&\dot{n}_{A}=D_{A}^{\mathcal{S}}\triangle_{\mathcal{S}}n_{A}-D_{B}\Big(\nabla n_{B}-\frac{\upsilon}{k_{B}T}n_{B}\nabla\sigma\Big)\cdot\boldsymbol{n}+F(n_{A},n_{C}),\\
&\dot{n}_{C}=D_{C}^{\mathcal{S}}\triangle_{\mathcal{S}}n_{C}+G(n_{A},n_{C}),
\end{aligned}\right\}\quad\text{on}\quad\partial\mathcal{R},
\end{aligned}\right\}\tag{1}$$

with $\hat{K}(\sigma)=K\exp\left(-\upsilon\sigma/(k_{B}T)\right)$ and $\sigma=\kappa(\operatorname{div}\boldsymbol{u}-\upsilon(n_{B}-n_{\mathrm{ref}}))$; where λ and G are the Lamé parameters, $\kappa=(3\lambda+2G)/3$ is the bulk modulus, n_{ref} is a reference concentration, υ is the molar volume, k_{B} is the Boltzmann constant, T is the temperature, $\boldsymbol{s}_{\mathrm{env}}$ is the traction exerted on $\partial\mathcal{R}$ by the environment, K is the adsorption constant, $\boldsymbol{u}$ is the displacement vector; n_{A}, n_{B}, and n_{C} represent the concentrations of constituents A, B, and C, respectively; D_{B} is the diffusion coefficient of constituent B on the bulk; $D_{A}^{\mathcal{S}}$ and $D_{C}^{\mathcal{S}}$ are the diffusion coefficients at the surface of constituents A and C, respectively; and, $F(n_{A},n_{C})$ and $G(n_{A},n_{C})$ are the source/sink terms that describe the kinetics of reactions. The symbols ∇, div, $\triangle$ and $\triangle_{\mathcal{S}}$ denote the gradient, divergence, Laplacian, and Laplace-Beltrami operators, respectively.

To obtain the equations we are going to use, we have followed the procedure discussed by the authors in (Duda *et al.*, 2023). The set of equations (Eq. (1)) is nondimensionalised using the relations $\hat{t}=t/t_{c}$, $\hat{\mathbf{x}}=\mathbf{x}/\ell_{c}$, $\hat{\boldsymbol{u}}=\boldsymbol{u}/\ell_{c}$, $a=n_{A}/\bar{n}_{A}$, $b=n_{B}/\bar{n}_{B}$, and $c=n_{C}/\bar{n}_{C}$, where t_{c} is a characteristic time, ℓ_{c} is a characteristic length, and $\bar{n}_{A}$, $\bar{n}_{B}$, and $\bar{n}_{C}$ are reference concentrations. Adopting the chemical scheme provided by Malevanets and Kapral (1997) to produce a kinetics of the Fitzhugh–Nagumo type, considering that $\hat{K}(\hat{\sigma})b\ll 1$ and introducing the following change of variables,

$$b=(c_{A}U+a_{0})/K,\qquad a=c_{A}u+a_{0}\qquad\text{and}\qquad c=c_{C}v+c_{0},\tag{2}$$

where a_{0}, c_{A}, c_{C} and c_{0} are constants that are related to the reactions rates. We arrive at the following set o equations[1]:

$$\left.\begin{aligned}
&\left.\begin{aligned}
&\operatorname{div}\big(\eta(\operatorname{div}\boldsymbol{u})\boldsymbol{I}+(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\top})-\hat{\kappa}\Omega(U-U_{\mathrm{ref}})\boldsymbol{I}\big)=\boldsymbol{0},\\
&\dot{U}=\operatorname{div}\Big(\alpha(1+\xi\hat{\kappa}\Omega(U+\zeta))\nabla U-\alpha\xi\hat{\kappa}(U+\zeta)\nabla(\operatorname{div}\boldsymbol{u})\Big),
\end{aligned}\right\}\quad\text{in}\quad\mathcal{B};\\
&\left.\begin{aligned}
&\big(\eta(\operatorname{div}\boldsymbol{u})\boldsymbol{I}+(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\top})-\hat{\kappa}\Omega(U-U_{\mathrm{ref}})\boldsymbol{I}\big)\boldsymbol{n}=\hat{\boldsymbol{s}}_{\mathrm{env}}\\
&(U+\zeta)\exp\Big(-\xi\hat{\kappa}(\operatorname{div}\boldsymbol{u}-\Omega(U-U_{\mathrm{ref}}))\Big)=u+\zeta,\\
&\dot{u}=d_{u}\triangle_{\mathcal{S}}u-\beta\Big((1+\xi\hat{\kappa}\Omega(U+\zeta))\nabla U-\xi\hat{\kappa}(U+\zeta)\nabla(\operatorname{div}\boldsymbol{u})\Big)\cdot\boldsymbol{n}+f(u,v),\\
&\dot{v}=d_{v}\triangle_{\mathcal{S}}v+g(u,v),
\end{aligned}\right\}\quad\text{on}\quad\partial\mathcal{B},
\end{aligned}\right\}\tag{3}$$

with $\eta=\lambda/G$, $\hat{\kappa}=\kappa/G$, $\hat{\boldsymbol{s}}_{\mathrm{env}}=\boldsymbol{s}_{\mathrm{env}}/G$, $\Omega=\upsilon\bar{n}_{B}c_{A}/K$, $\xi=G\upsilon/(k_{B}T)$, $\zeta=a_{0}/c_{A}$, $d_{u}=D_{A}^{\mathcal{S}}t_{c}/\ell_{c}^{2}$, $d_{v}=D_{C}^{\mathcal{S}}t_{c}/\ell_{c}^{2}$, $\alpha=D_{B}t_{c}/\ell_{c}^{2}$ and $\beta=D_{B}t_{c}\bar{n}_{B}/(\ell_{c}\bar{n}_{A}K)$. Lastly, terms f and g are defined by,

$$f(u,v)=-u^{3}+u-v\qquad\text{and}\qquad g(u,v)=\epsilon(u-\gamma v-\lambda),\tag{4}$$

which describes the Fitzhugh–Nagumo kinetics, where ϵ, γ, and λ are model parameters that control the kinetics displayed by the system. For convenience, we have dropped the $\hat{(\cdot)}$ to denote dimensionless variables and the symbols ∇, div, $\triangle$, and $\triangle_{\mathcal{S}}$ denote the dimensionless version of the originally introduced operators.

[1] A detailed derivation of the reaction terms for the transport equations on the surface can be found in Duda *et al.* (2023).

3. NUMERICAL IMPLEMENTATION

The set of equations (Eq. (3)) was implemented and solved numerically using the finite element software COMSOL Multiphysics® v. 5.6 (2020). The equations for U, u, and v, were implemented in the strong form using the general form PDE module for the bulk quantity and the boundary general form PDE module for the surface quantities. The coupling between bulk diffusion and mechanical deformation was made through an analogy with the thermal expansion subnode inside the solid mechanics module. The fields (U, u, v) were approximated using piece-wise linear functions while the displacement vector field, $\boldsymbol{u}$, was approximated by piece-wise quadratic functions since the flux contains second-order derivatives of the displacement vector field components. For time discretization, the second-order backward differentiation formulation (BDF) was used. At each step time, the system of nonlinear algebraic equations was solved using the direct solver PARDISO in a fully coupled scheme. In this study, the computational domain was a unit sphere discretized by a mesh comprising 22,916 tetrahedral elements in the bulk and 5,280 triangular elements on the surface.

4. RESULTS AND DISCUSSION

For this study, we have adopted the same parameters used by Krause *et al.* (2018) for the Fitzhugh–Nagumo model, in which $d_u = 1/400$, $d_v = 1/20$, $\epsilon = 1.2$, $\gamma\epsilon = 1.1$, and $\lambda = 0$. To produce the chemical patterns on the surface of the sphere, a perturbation of the homogeneous steady state is necessary. For this reason, we have adopted the method of Krause *et al.* (2018), in which a random initial condition, generated from a normal distribution with mean zero and standard deviation of 10^{-3}, was used as initial condition on the surface variables.

To study the influence of the mechanical effect on the pattern formation, a pressure load, $\hat{\boldsymbol{s}}_{\text{env}} = -\hat{p}_{\text{env}}\boldsymbol{n}$, was applied to the surface of the sphere. In Fig. 1, we show the distribution of the quantities U and u on the surface of the sphere for different environment pressures, $\hat{p}_{\text{env}}$. We can notice that, as the pressure increases, the pattern is suppressed on the bulk variable, U, while it remains on the surface variable, u. This suggests that, for this problem, mechanics has a stabilizing effect on the pattern formation of the bulk variable. This outcome can be expected, since the gradient of the mean stress appears to be low within the sphere, and high pressure values tend to suppress bulk-surface interaction through the sorption processes. During this analysis, we also noticed that material mechanical properties, such as the Poisson ratio, have no significant effect on the pattern formation. This observation is in agreement with the work of Brinkmann *et al.* (2018).

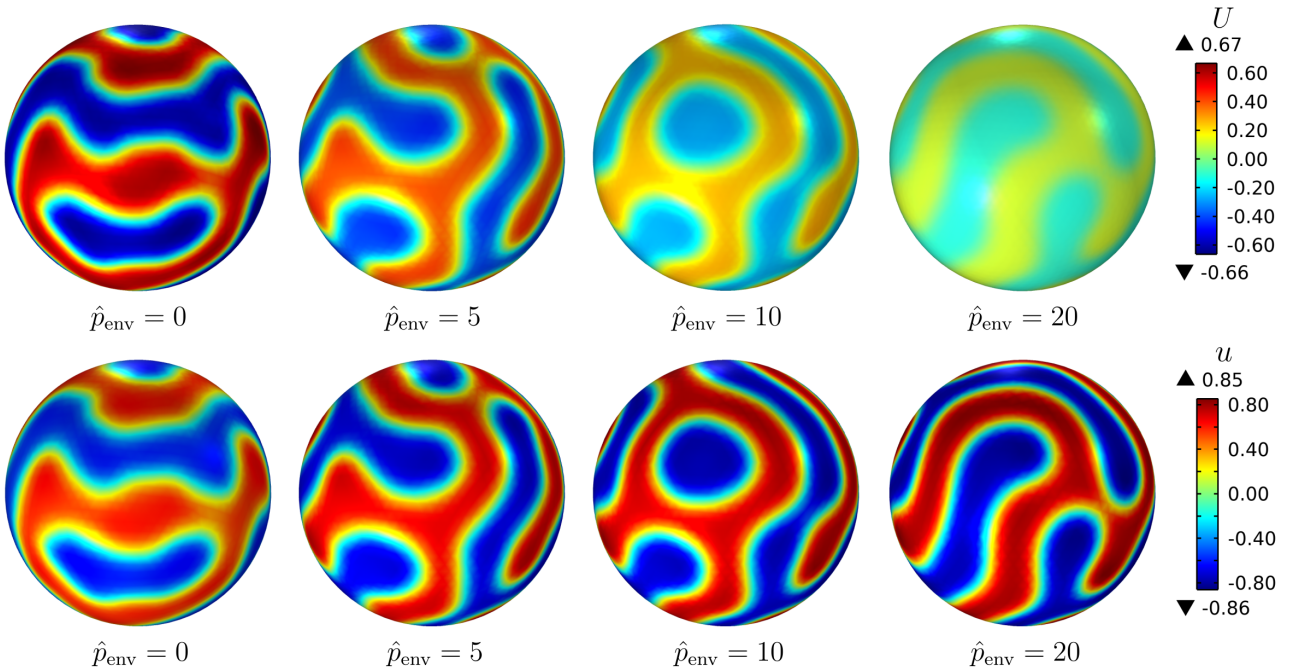

Figure 1: Pattern formation at $t = 100$ starting from a random initial condition for $d_u = 1/400$, $d_v = 1/20$, $\epsilon = 1.2$, $\gamma\epsilon = 1.1$, $\lambda = 0$, $\alpha = 1$, $\beta = 0.05$, $\eta = 1.5$, $\Omega = 0.1$, $\xi = 0.1$, $\zeta = 0$, and different values of $\hat{p}_{\text{env}}$.

5. CONCLUDING REMARKS

We have studied the phenomenon of chemical pattern formation on the surface of a deformable body. The model was implemented and solved using the finite element software COMSOL Multiphysics. The numerical results suggest that a mechanical load, such as a pressure exerted by the environment, can stabilize the surface suppressing chemical pattern

formation in the bulk variable. In a future investigation, the mechanical effect on pattern formation will be addressed through a linear stability analysis.

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