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Effect of material anisotropy on the fingering instability in reverse smoldering combustion

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Abstract

It is well known from experiments that a sample of thin porous material burning against an oxidizing air under microgravity exhibits various finger-like char patterns. The patterns are classified into three distinct types depending on the oxidizer flow rate. (I) Sparse fingers; (II) tip-splitting fingers; (III) connected front. We presently extend our modeling strategy based on the homogenization approach, which has been applied for the case of isotropic porous media, to analyze the pattern behavior on anisotropic porous media. In order to understand the characteristic features of the patterns based on the influence of the local structure, we simply rely on fixed anisotropic two-dimensional geometries representative of the microstructure of interest. Thus, we illustrate numerically the consequence of the material anisotropy on the fingering patterns based on effective diffusion tensors calculated using the homogenization method and the mechanism of thermal-diffusion instability. Besides revealing new insights on the experimental observations, our numerical results show that material anisotropy can influence the uniformity on the patterns, but the distinct fingering regimes are independent of the local microstructure of materials. This effect is consistent with the qualitative experimental findings from Zik and Moses (1999).

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1. Introduction

Smoldering describes a slow, low temperature and non-flaming mode of combustion. Smolder waves basically proceed as a self-sustaining reaction front that propagates on the surface of a solid porous sample, which reacts with an oxidizer gas infiltrating its pores. The direction of flow of the oxidizer gas relative to the direction of propagation of the reaction front can be classified into distinct regimes and has been a subject of practical interest due to the characteristic features exhibited by each of the regimes. For a detailed discussion on the distinct smoldering regimes, we refer to [2–5, e.g.]. In this paper, we focus on the smoldering regime in which the oxidizer gas flows in a direction opposite to the direction of the reaction front. The literature, in this regime, is referred to as the reverse smoldering regime. Our interest in reverse smolder fronts is motivated by the experimental work detailed in [1]. It is shown in [1] that the reverse regime promotes the destabilization of a combustion front. The instability of the combustion front arises due to a destabilizing effect of oxidizer transport [1,6,7], which manifest in the form of finger-like char patterns in the absence of natural convection. Similar phenomenological finger-like patterns have been observed aboard a spacecraft [8]. The fingering instability can be grouped into three distinct fingering regimes based on the velocity of the oxidizer flow: the sparse fingers, tip-splitting fingers, and connected front (see Fig. 1). According to [1], the experiment is setup in such a way as to maintain uniform flow, ignition and material. Thus, the fingering patterns are uniform, in the sense that the direction of propagation of the fingers may not necessarily be in the direction of the oxidizer flow. The patterns may manifest distinct forms of nonuniformities as described in [1]. Here, we illustrate in Fig. 2(b) some forms of nonuniformity. Thus, the experimental observations lead to the basic question of whether the local microstructure of a porous material can influence the fingering behavior under conditions of uniform flow and ignition. This is the main objective of the present paper. Recently, the macroscopic description of the smoldering combustion problem has been derived from a basic pore scale description in an isotropic porous medium [9] by using the homogenization theory based on periodic structures [14–18] and two-scale convergence methods [19,20]. The main assumptions related to the method by homogenization, the physics of the phenomenon of interest at the pore scale and the main results deduced from the upscaling procedure are briefly recalled in Section 2. In Section 3, we analyze numerically the properties of the effective diffusion tensors derived through homogenization method for different angular orientations of a centered elliptical inclusion. The correct forms of the anisotropic diffusion tensors are also deduced. In Section 4, the tensorial properties of the elliptical geometry are then used to formulate distinct functional forms of macroscopic system of equations for smoldering combustion of anisotropic porous media. The behavior of the fingering patterns is analyzed numerically for simplified anisotropic media having distinct angular orientations of the elliptical inclusions in Section 5. For each realization based on an angular orientation, the medium is assumed to be uniformly periodic with fixed volume fraction of the elliptical inclusion. We point out that the choice of elliptical microstructures used in this

![Fig. 1. Spatial profiles of two-dimensional fingering (char) patterns of a filter paper sample, observed experimentally in a Hele-Shaw cell. The char propagation is from bottom to top; Ignition is initiated at the bottom and oxidizer gas is passed from the top, in a typical counterflow configuration. The char is identified by the dark finger-like patterns, and the light shades are the quenched part of the flame, which separates regions of burned parts from unburned parts. (a) Connected front which manifests at high flux velocity; the fingers are connected. (b) Tip-splitting regime marked by splitting of sole fingers at the tips; it is observed at a moderate flux velocity. (c) Sparse fingers, which manifest at a relatively low flux velocity; the fingers are more distinct from each other and the tips do not split. The snapshots are courtesy of E. Moses (Weizmann Institute of Science).](image-url)
study is basically for simplicity; more complex microstructures are also applicable.

2. Microscopic modeling

2.1. Microscopic description

In a previous work [9], we analyzed the problem discussed presently for an isotropic medium by using the method of homogenization for periodic structures. This method allows to establish the general form of the macroscopic law for a smoldering combustion scenario. The homogenization process involves starting from a basic pore scale description of the microstructure and the physics of the phenomenon of interest, in which one assumes that the porous sample and the physical quantities of interest are periodic. Let us consider the microscopic zoom of a thin porous sample of Whatman filter paper depicted in Fig. 3. It is clear that the material consists of complex arrangement of fibers, which are not uniformly arranged, but intrinsically oriented in a complex manner throughout the medium. By identifying distinct angular orientations of the fiber, the homogenization method for periodic structures can then be applied in such a way that for each fiber orientation one assumes that the medium consist of a uniformly periodic arrangement of a typical representative elementary volume (REV). A realization of the uniformly periodic medium is illustrated in Fig. 4. The validity of the method requires that the condition of scale separation is satisfied, i.e. the ratio of the characteristic length, \( l_c \), of the REV to the characteristic length, \( L_c \), of the macroscopic sample is a small parameter represented by \( \varepsilon = l_c / L_c \). In the sequel, we briefly recall the deductive strategy behind the models derived in [9]. The REV of the porous domain (see Fig. 4) consists of two parts: a solid fiber filled part, \( Y_s \), with smooth boundary \( \partial Y_s \) and a gas filled part, \( Y_g \), so that the ensemble of the solid phase occupies a volume \( \Omega_s \). An oxidizer gas infiltrates a pore space of total volume \( \Omega_g \) left by the fibrous solid phase. The interphase between the solid and the gaseous phase is denoted by \( \Gamma \). We neglect the hydrodynamic aspects of the flow [21] and assume instead that the flow in the porous medium is a constant laminar field \( \mathbf{u} \) given by

\[
\mathbf{u}(x) = \begin{cases} 
0, & x \in \Omega_s \\
\mathbf{v}, & x \in \Omega_g 
\end{cases}
\] (1)

---

Fig. 2. Schematic diagram of uniform and nonuniform finger formation. (a) Uniform fingering pattern; (b) nonuniform fingering pattern; the fingers are directed toward a boundary and can also emerge in clusters. The arrows adjacent to the fingers indicate direction of propagation.

Fig. 3. Microscopic view of a filter paper sample showing complex arrangement of the cellulosic structure at different angular orientations. Each orientation is represented by a representative elementary volume with elliptical inclusion. The snapshots are courtesy of N. J. Suematsu (Meiji Institute for Advanced Study of Mathematical Sciences).

Fig. 4. Scheme of the periodic medium realized from a given orientation of the inclusion as illustrated in Fig. 3.
where (1) implies the restriction of convection to the gas phase \( \Omega_g \).

At the pore scale, the heat balance laws in the gas and solid phases are given by

\[
\frac{\partial T_g}{\partial t} + C_g \mathbf{u} \cdot \nabla T_g - \nabla \cdot (\lambda_g \nabla T_g) = 0, \quad \mathbf{x} \in \Omega_g, \quad t > 0, \tag{2a}
\]

\[
\frac{\partial T_s}{\partial t} - \nabla \cdot (\lambda_s \nabla T_s) = 0, \quad \mathbf{x} \in \Omega_s, \quad t > 0, \tag{2b}
\]

where \( C_i = \rho_i c_i \) is the volumetric heat capacity of phase \( i \), \( \lambda_i \) is the heat conductivity and \( T_i \) is the temperature. Since the reaction between the phases is purely heterogeneous and takes place on the surface of the solid, the transport equations are coupled at the pore boundary \( \Gamma \). Also, the mass balance law for the gaseous oxidizer infiltrating the pore volume \( \Omega_q \) is given by

\[
\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C - \nabla \cdot (D \nabla C) = 0, \quad \mathbf{x} \in \Omega_q, \quad t > 0, \tag{3}
\]

where \( C \) is the concentration and \( D \) is the diffusion coefficient, assumed here to be constant. The chemical process in the combustion regime assumes the following form

Solid fuel + \( \text{O}_2 \) \rightarrow solid product + gas product + heat. \tag{4}

For an oxygen-limited reaction, the reaction rate \( W \) is given by a one-step first order reaction with respect to the deficient gaseous reactant. The temperature-dependent rate coefficient \( k \) is governed by the Arrhenius’ law, viz.

\[
k(T_s) = A \exp \left( -\frac{T_a}{T_s} \right). \tag{5}
\]

Here, \( A \) is the pre-exponential factor, \( T_s \) is the activation temperature of the reaction. The reaction at the surface \( \Gamma \) is given by the following boundary conditions

\[
(\lambda_g \nabla T_g - \lambda_s \nabla T_s) \cdot \mathbf{n} = QW(T_s, C), \quad \mathbf{x} \in \Gamma, \quad t > 0, \quad T_g = T_s, \quad \mathbf{x} \in \Gamma, \quad t > 0, \quad D \nabla C \cdot \mathbf{n} = -W(T_s, C), \quad \mathbf{x} \in \Gamma, \quad t > 0, \tag{6a-6c}
\]

where \( \mathbf{n} \) is the outward unit normal vector which points in a direction outside \( Y_g \) and \( Q \) is the heat release. \( W(T, C) \) is given by

\[
W(T, C) = A C \exp \left( -\frac{T_a}{T_s} \right). \tag{7}
\]

Additionally, the char product \( R \), is given by

\[
\frac{\partial R}{\partial t} = W(T, C), \quad \mathbf{x} \in \Gamma, \quad t > 0. \tag{8}
\]

So far, the governing equations together with the associated interior coupling conditions have been described. Additionally, the following boundary conditions are prescribed at the exterior boundaries:

\subsection*{2.1.1. Diffusive thermal insulation conditions}

\[
\mathbf{n} \cdot \nabla T_i = 0, \quad \mathbf{n} \cdot \nabla C = 0, \quad \partial \Omega \setminus (\{ \mathbf{x} = 0 \} \cup \{ \mathbf{x} = a_i \}), \quad t > 0. \tag{9}
\]

\subsection*{2.1.2. Upstream/Downstream boundary conditions}

\[
T_i = T_u, \quad C = C_u, \quad \{ \mathbf{x} = 0 \} \cap \partial \Omega, \quad t > 0, \tag{10a}
\]

\[
\mathbf{n} \cdot \nabla T_i = 0, \quad \mathbf{n} \cdot \nabla C = 0, \quad \{ \mathbf{x} = a_i \} \cap \partial \Omega, \quad t > 0. \tag{10b}
\]

Here, \( u \) denotes the unburnt or initial values, and \( i = g \). The problem is further supplemented with appropriate initial conditions in order to completely describe the problem.

\subsection*{2.2. Parameter estimation}

The form of rescaling introduced in [9] is used to establish the correct estimates for the dimensionless parameters in the system of Eqs. (2)-(6). By using the macroscopic viewpoint [22], the characteristic length of the domain is \( L \). Thus, the dimensionless form of (2)-(6) introduces the following dimensionless numbers:

\[
p_e := \frac{L \gamma_c}{D_c} = \frac{t_0}{\tau_c} \quad \text{(Péclet number)}, \tag{11a}
\]

\[
Le_g := \frac{r_c}{D_c} \quad \text{(gas-phase Lewis number)}, \tag{11b}
\]

\[
Da := \frac{L \alpha_c}{D_c} = \frac{t_D}{\tau_c} \quad \text{(Damköhler number)}. \tag{11c}
\]

We take the characteristic time of diffusion in the subdomain, \( \Omega_g \), as the characteristic time of the observation at the macroscopic scale, i.e., \( t_c = t_0 \). Rewriting (2) in terms of the introduced dimensionless numbers leads to following:

\[
\frac{\partial T_g}{\partial t} + C_g p_e \mathbf{v} \cdot \nabla T_g - Le_g \mathbf{v} \cdot (\lambda_g \nabla T_g) = 0 \tag{12a}
\]

\[
\frac{\partial T_s}{\partial t} - \mathbf{v} \cdot (\lambda_s \nabla T_s) = 0. \tag{12b}
\]

After introducing some simplifications, the corresponding inter-phase conditions are

\[
\mathbf{n} \cdot (\lambda_e \nabla T_g - \mathbf{v} \cdot \lambda_s \nabla T_s) = Da Q' W'(T_s, C'), \tag{12a}
\]

\[
T_g = T_s, \quad T_s = T_0, \tag{12b}
\]

with

\[
W'(T_s, C') = A' C' \exp \left( -\frac{T_a}{T_s(T_a - T_b) + T_b} \right), \tag{13}
\]

where \( \lambda_e \) is the gas phase thermal diffusivity. The ratio of heat capacities in the material is denoted as \( m \) and the corresponding ratio of heat conductivities is \( \kappa \). Further, we define the temperature of combustion product, \( T_b := T_0 + Q_c/C_c/p_{e,g} \). The global characteristic time scales above are defined as follows:

\[
t_D := \frac{L^2}{D_c}, \quad t_a := \frac{L}{u_c}, \quad t_c := \frac{L_c}{A_c}, \quad t_b := \frac{R_i}{C_c}, \tag{13}
\]

where \( t_a \) is the characteristic global diffusion time scale, \( t_b \) is the characteristic global advection time scale, \( t_c \) and \( t_b \) are respectively the characteristic time of gas reaction and the characteristic time of combustion product. We point out that all quantities subscripted with “c” define some characteristic macroscopic quantities. Similarly, the mass concentration of oxygen in dimensionless form is:

\[
\frac{L^2}{D_c} \frac{\partial C}{\partial t} + u_c \frac{\partial C}{\partial x} = -\mathbf{v} \cdot \nabla C - \nabla \cdot (D \nabla C) = 0, \quad \{ \mathbf{x} = 0 \}, \tag{13}
\]

where

\[
\tau_c := \frac{L^2}{D_c} \quad \text{the ratio of global characteristic transport times}. \tag{14}
\]

With the choice of characteristic time of diffusion \( t_D \), we introduce the Péclet number in (13) to get

\[
p_c \frac{\partial C}{\partial t} + p_{e,g} \mathbf{v} \cdot \nabla C - \nabla \cdot (D \nabla C) = 0. \tag{14}
\]

According to [9], all the dimensionless quantities are estimated by a unit order of magnitude, i.e. \( O(1) \), except \( Da = O(\varepsilon) \). This is particularly true for problems in which the thermal conductivity of the inclusions are of the same order of magnitude to that of the gaseous phase.
2.3. Asymptotic analysis

The homogenization method for periodic structures is used to obtain the macroscopic system of equations. The first step is to assume that solutions to the unknown fields in (2)–(6) follow a two-scale asymptotic expansion, namely:

$$
\begin{align*}
T_i(x, t) &= T_i^{(0)}(x, y, t) + \varepsilon T_i^{(1)}(x, y, t) + \cdots, \\
C(x, t) &= C^{(0)}(x, y, t) + \varepsilon C^{(1)}(x, y, t) + \cdots, \\
\mathcal{R}(x, t) &= \mathcal{R}^{(0)}(x, y, t) + \varepsilon \mathcal{R}^{(1)}(x, y, t) + \cdots.
\end{align*}
$$

(15)

where $T_i^{(n)}, C^{(n)}, \mathcal{R}^{(n)}, n = 1, 2, \ldots, i = \{g, s\}$ are $Y$ periodic in $y$ with $y = \frac{x}{\varepsilon}$. Due to the scale separation $\{x, \frac{x}{\varepsilon}\}$, the unknowns in (15) are functions of three variables: $x, y$ and $t$. Consequently, we transform the derivatives through the chain rule:

$$
\nabla_x = \nabla_y + \frac{1}{\varepsilon} \nabla_y.
$$

(16)

Applying the expansions (15), using (16) and collecting terms with the same powers of $\varepsilon$, successive boundary value problems are obtained.

2.4. Macroscopic description

From the successive boundary value problems, it can be shown by the homogenization process [9] that the first terms of (15) depend only on the macroscopic variable $x$ and the second terms of the first two expansions are defined by

$$
\begin{align*}
T_i^{(1)}(x, y, t) &= \sum_{j=1}^{2} \frac{\partial T_j^{(0)}}{\partial x}(x, t)M_j(y), \\
C^{(1)}(x, y, t) &= \sum_{j=1}^{2} \frac{\partial C_j^{(0)}}{\partial x}(x, t)N_j(y),
\end{align*}
$$

(17)

(18)

where $M_j, N_j, i = 1, 2,$ are the solutions of the so-called cell problems given respective by:

$$
\begin{align*}
-dv_y (\lambda_y (e_y + \nabla M_j)) &= 0 \quad \text{in} \ Y_y, \\
-dv_y (\lambda_y (e_y + \nabla M_j)) &= 0 \quad \text{in} \ Y_y, \\
-\lambda_y (e_y + \nabla M_j) \cdot n &= \lambda_y (e_y + \nabla M_j) \cdot n \quad \text{in} \ \partial Y_y, \\
M_j &= \text{Y-periodic}, \\
-dv_y (D(e_y + \nabla N_j)) &= 0 \quad \text{in} \ Y_y, \\
-D(e_y + \nabla N_j) \cdot n &= 0 \quad \text{in} \ \partial Y_y, \\
N_j &= \text{Y-periodic}.
\end{align*}
$$

(19)

Additionally, the vector valued functions $M$ and $N$ satisfy the condition of zero mean, i.e.

$$
\langle \rho \rangle = \frac{1}{|V|} \sum_{i \in \{g, s\}} \int_{Y_y} \rho_i dY = 0.
$$

(20)

Finally, the macroscopic mass and heat transport balance equations that describe the averaged limit behavior of the microscopic descriptions (2)–(6) are, respectively:

$$
\begin{align*}
\langle \phi \frac{\partial C_{\text{eff}}}{\partial x} + \phi \nabla \cdot \nabla C^{(0)} - \nabla \cdot (D^{\text{eff}} \nabla C^{(0)}) \rangle &= -\phi^s W(T^{(0)}, C^{(0)}), \\
\langle \phi c_{\text{eff}} \frac{\partial T_{\text{eff}}}{\partial x} - \nabla \cdot (\lambda_{\text{eff}} \nabla T^{(0)}) + \phi c_s \nabla \cdot \nabla T^{(0)} \rangle &= \phi^s Q(W(T^{(0)}, C^{(0)}), \\
\langle \frac{\partial p_{\text{eff}}}{\partial x} \rangle &= W(T^{(0)}, C^{(0)}).
\end{align*}
$$

(21)

The effective thermal conductivity tensor is defined by:

$$
\lambda_{\text{eff}}^{ij} = \frac{1}{|V|} \left( \int_{Y_y} \lambda_y \left( I_y + \frac{\partial M_j}{\partial y} \right) dY + \int_{Y_y} \lambda_y \left( I_y + \frac{\partial M_j}{\partial y} \right) dY \right),
$$

(22)

$i, j = 1, 2$. Also, the effective volumetric heat capacity is defined by:

$$
C_{\text{eff}} = \frac{1}{|V|} \left( \int_{Y_y} C_g dY + \int_{Y_y} C_s dY \right) = \phi C_s + (1 - \phi) C_g.
$$

(23)

The effective diffusion tensor $D_{\text{eff}}$ is defined by:

$$
D_{\text{eff}}^{ij} = \frac{D}{|V|} \int_{Y_y} \left( I_y + \frac{\partial N_j}{\partial y} \right) dY.
$$

(24)

Here, $\phi^t$ and $\phi$ are respectively the tortuosity and porosity factors.

3. Analysis of the effective diffusion tensors

3.1. Computation of the effective diffusion tensor

The effective diffusion tensors are computed numerically by using the derived formula (22) and (24). These calculations require solving the cell problems (19) posed on a single period. The main objective of this subsection is to analyze the sensitivity of the effective tensors to angular orientation of the elliptical inclusion. In other contexts, however, it is also possible to carry out the analysis by considering variations in the aspect ratio of the REV, and hence the volume fraction of the inclusion [23–25]. The numerical simulations are done in a single periodic REV for thermal conductivity values $\lambda_y = 2.38 \times 10^{-4}$ W/cm K and $\lambda_s = 7 \times 10^{-4}$ W/cm K, corresponding respectively to the gaseous part and the solid part. The conductivity values correspond to a moderately thermal conducting medium, in which the conductivities of the phases are comparable and hence the underlying homogenization problem can be estimated by an order of magnitude, in terms of the scale parameter $\varepsilon$, equal to $O(1)$ (cf. [9,22,26,27]). The gaseous phase molecular diffusion constant is given by $D^\text{mol} = 0.25$ cm$^2$/s.

3.2. Anisotropy of the effective diffusion tensors

Anisotropy effect is a property of nonuniform porous materials. It means that the direction of flow in such a medium is not simply the direction of the gradient of the transport processes. There are a couple of literature dealing with anisotropy, which is induced through effective diffusion properties [28–30]. In order to mimic a physical material that exhibits anisotropy, we performed numerical simulations for a geometry with a centered elliptical inclusion. We take, for example, a typical elliptical inclusion having an A-semiaxes of $a = 0.3$ and a B-semiaxes of $b = 0.48$ Thus, the volume fraction of the inclusion is fixed to $\eta = rab = 0.45$. We also use the same value
of $V_s$ for a test problem in an isotropic geometry with circular inclusion simply for the purpose of comparison with the problems on the anisotropic geometry. The REV has a unit dimension, which makes sense for an REV having an aspect ratio of 1 [24]. For our purpose, the angular orientation, $\theta$, of the elliptical inclusion is varied clock-wise in steps of 15° about its major axis (see Fig. 5). We chose a range $\theta = 0°; 15°;...; 180°$. It should be noted that within the given range, the resulting tensors have distinct properties. The form of the tensors can be grouped into two categories; diagonally anisotropic and symmetrically anisotropic. The derived effective tensors on the right half of the range, i.e. $\theta = 105°; 120°;...$, have negative off-diagonal entries which, for a generic tensor

$$\lambda = \begin{pmatrix} a & b \\ c & d \end{pmatrix},$$

defined on the left half of the range, tensors on the right half are obtained through the following representation

$$\lambda^\theta = \begin{pmatrix} a & \mu b \\ \mu c & d \end{pmatrix},$$

where $\mu = \text{sgn}\cos\theta$. Fig. 7 shows the solutions of the local boundary value problems (19) for different values of $\theta$. In Fig. 7(a) and (b), the cell function $M$ is given in the $e_1$ axis and for $\theta = 45°$ and $\theta = 120°$ respectively, while in Fig. 7(c) and (d) the cell function $N$ is depicted for $\theta = 60°$ in the $e_1$ axis and $\theta = 0°$ in the $e_2$ axis respectively. The results of numerical calculation showed that the entries of the effective thermal conductivity and mass diffusivity tensors depend on $\theta$. They lead to different forms of anisotropic tensors. At $\theta = 0°; 90°; 180°$, the effective diffusion tensors are diagonally anisotropic (DA), as shown in Fig. 6(a) and (b). In between these points, the tensors are basically symmetrically anisotropic, with a peak magnitude of the anisotropy occurring at $\theta = 45°$, respectively $\theta = 135°$. Further, we see that the principal entries of the effective tensors coincide at $\theta = 45°$ (cf. $\theta = 135°$). These values are also

---

Fig. 6. Evolution of the effective diffusion tensors as a function of $\theta$. (a) Evolution of the entries of effective thermal conductivity tensor, $\lambda^{\text{th}}$, with $\theta$; (b) evolution of the entries of the effective mass diffusivity tensor, $D^{\text{th}}$, with $\theta$. The points marked with DA are diagonally anisotropic with respect to $\theta$. In both cases, the diffusion constants for the isotropic geometry with $V_s = 0.45$ coincides with the principal entries of the tensors at $\theta = 45°, 135°$

---

Fig. 7. Solution of the local boundary value problem (19) with centered elliptical inclusion.
consistent with the values of the effective thermal conductivity, $\lambda_{\text{eff}}$, and mass diffusivity, $D_{\text{eff}}$, constants for the isotropic geometry with the same value volume fraction $V_s$ of a circular inclusion. According to [31], It can be shown that the numerical values of the principal entries of the tensors are consistent with the Voigt–Reuss bounds [22,32]. The principal entries of the tensor, $D_{\text{eff}}$, are more closer to the Voigt upper bound whereas those of $\lambda_{\text{eff}}$ are closer to the Reuss lower bound. However, the off-diagonal entries are farther below. This result implies that the Voigt–Reuss bounds are consistent with the principal direction of flow in both cases, even though the off-diagonal directions may not be insignificant. The entries of the diffusion tensors on the right half of the range $\theta = 105^\circ, 120^\circ, \ldots, 165^\circ$, are reflections of the entries on the left half, since their magnitudes are the same.

4. Macroscopic modeling

4.1. Macroscopic model for general anisotropic tensors

The numerical results and analysis of the effective diffusion tensors presented in the previous section showed distinct tensorial properties of the elliptical geometry. Thus, the form of the macroscopic system of equations can now be studied in terms of the properties of these tensors. In this subsection, we begin by describing the general form of the anisotropic system of equations, which serves as a generalization for both the isotropic and subsequent anisotropic systems to be discussed. We recall that the main objective of this paper is to analyze nonuniformity on the fingering patterns based on anisotropy effect induced through the effective diffusion tensors. Since the tensors possess distinct characteristic properties for each value of $\theta$, we expect that they exhibit different magnitudes of effect on the fingering patterns. Thus, it makes sense to formulate distinct macroscopic equations based on the tensorial properties of the effective diffusion tensors. The general form of the anisotropic system of balance equations is given by

$$
\frac{\partial \tilde{\rho}}{\partial t} = \tilde{\rho} \frac{\partial \tilde{\rho}}{\partial x} + \tilde{\rho} \frac{\partial ^2 \tilde{T}}{\partial x^2} + \tilde{\rho} \frac{\partial ^2 \tilde{T}}{\partial y^2} + \tilde{\rho} \frac{\partial ^2 \tilde{T}}{\partial z^2} + \psi \hat{Q}(T,C),
$$

$$
\frac{\partial C}{\partial t} + \tilde{\rho} \frac{\partial \tilde{C}}{\partial x} + \tilde{\rho} \frac{\partial \tilde{C}}{\partial y} + \tilde{\rho} \frac{\partial \tilde{C}}{\partial z} = D_{\text{eff}} \left( \frac{\partial ^2 \tilde{C}}{\partial x^2} + \frac{\partial ^2 \tilde{C}}{\partial y^2} + \frac{\partial ^2 \tilde{C}}{\partial z^2} \right),
$$

$$
\frac{\partial R}{\partial t} = H_R(T,C),
$$

where

$$
W = ANC \exp\{N(1 - 1/T)\}.
$$

In (29), $Pe = \mu_{\text{th}} / \lambda_{\text{eff}}$ is the effective Péclet number. $H_R = C_\gamma / \phi^2 R_q$ and $\Lambda = C_\gamma / C_{\text{eff}}$ represent dimensionless coefficients. The anisotropy indicators are represented by

$$
\gamma = \frac{\lambda_{xy}}{\lambda_{xx}}, \quad \mu_x = \frac{\lambda_{xy}}{\lambda_{xx}}, \quad \mu_y = \frac{\lambda_{xy}}{\lambda_{yy}}, \quad \omega = \frac{\lambda_{xy}}{\lambda_{yy}},
$$

where $\omega$, $\mu_x$, and $\mu_y$ denote respectively the longitudinal and transverse Lewis numbers, whereas $\omega_0$, $\mu_0$, and $\mu_0$ are the corresponding off-diagonal entries. In matrix notation (cf. (25) and (26)), the form of the anisotropic tensors with varying angular orientation, $\theta$, can be represented by

$$
\lambda = \left( \begin{array}{ccc} 1 - \omega_0 & \mu_0 & 0 \\ \mu_0 & 1 - \mu_0 & 0 \\ 0 & 0 & 1 - \omega \end{array} \right), \quad D = \left( \begin{array}{ccc} 1 - \omega_0 & \mu_0 & 0 \\ \mu_0 & 1 - \mu_0 & 0 \\ 0 & 0 & 1 - \omega \end{array} \right),
$$

where $\omega = \sin \theta \cos \theta$, with $\theta = 0^\circ, 15^\circ, \ldots, 165^\circ, 180^\circ$.

4.3. Macroscopic model for diagonally anisotropic tensors

From the numerical results of the preceding section, the macroscopic equations for diagonally anisotropic tensors can be realized by setting the off-diagonal entries of the tensors to zero, i.e.

$$
\lambda_{xy} = \mu_x = \mu_y = 0, \quad D_{xy} = D_{yx} = 0.
$$

The dimensionless system of equations, in this case, follows accordingly from (29). The anisotropy effect in the system is indicated by the introduction of an anisotropy indicator, $\gamma$. It generalizes the anisotropy in the system, i.e. we assume $\gamma$ to be such that we set

$$
\psi \gamma^{-1} := \gamma \psi^{-1}.
$$

Table 1

<table>
<thead>
<tr>
<th>Physico-chemical parameters</th>
<th>Unit</th>
<th>Gas</th>
<th>Solid</th>
<th>Other</th>
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<td>$c_p$</td>
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<td>–</td>
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<tr>
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<td>540</td>
<td>–</td>
</tr>
<tr>
<td>$D$</td>
<td>m(^2)s(^{-1})</td>
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<td>–</td>
<td>–</td>
</tr>
<tr>
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<td>0.07</td>
<td>14432</td>
</tr>
<tr>
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<td>11200</td>
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</tr>
<tr>
<td>$Q$</td>
<td>Jmol(^{-1})</td>
<td>–</td>
<td>–</td>
<td>2 \times 10(^6)</td>
</tr>
<tr>
<td>$A$</td>
<td>s(^{-1})</td>
<td>–</td>
<td>–</td>
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</table>
Fig. 8. Evolution of the entries of the anisotropic tensors with $\theta$. (a) Evolution of the effective Lewis numbers with $\theta$; (b) evolution of the anisotropic indicators $\mu$ and $\gamma$ with $\theta$.

Fig. 9. Evolution of char pattern for various values of $\theta$. (a) $\theta = 0^\circ$. The pattern is finger-like and sparse; (b) $\theta = 90^\circ$. The pattern is cusp-like and connected; (c) $\theta = 30^\circ$; (d) $\theta = 60^\circ$. In (c)–(d), the pattern is nonuniform and directed toward the lateral boundary. In the computations, $N = 1.8$, $Pe = 10$, except for (a) $Pe = 1$. 
The consideration here makes sense, since the primary control parameter for the pattern behavior is the Lewis number, and thermal anisotropy effect dominates in the considered system; hence, we treat \( \frac{L}{C_0} \) in terms of \( \frac{L}{C_0} \).

### 4.4. Macroscopic model for symmetrically anisotropic tensors

The macroscopic equations for the case of symmetrically anisotropic diffusion tensors can be realized from the general anisotropic Eq. (27) by setting the off-diagonal entries of the anisotropic tensors to:

\[
\begin{align*}
    \lambda_{yx}^{\text{eff}} &= \lambda_{xy}^{\text{eff}}, \\
    \phi_{yx}^{\text{eff}} &= \phi_{xy}^{\text{eff}},
\end{align*}
\]

such that the system of equations corresponding to (33) can be rewritten in dimensionless form as given by the following matrix notation:

\[
\lambda = \begin{pmatrix} 1 & \phi \mu / \gamma \\ \phi \mu / \gamma & \phi \mu / \gamma \end{pmatrix}, \\
D = \begin{pmatrix} \phi \mu / \gamma & \phi \mu / \gamma \\ \phi \mu / \gamma & \phi \mu / \gamma \end{pmatrix},
\]

where \( \mu \) and \( \gamma \) are the anisotropy indicators. The pattern behavior can also be characterized by the ratio, \( \sigma = \psi_{L} / \psi_{T} \). For example, if \( \sigma \ll 1 \), the pattern is sparse and if \( \sigma \gg 1 \), the pattern is more cusp-like and connected.

### 5. Results and discussion

#### 5.1. Fingering behavior based on anisotropic effective tensors

In the previous section, we have derived the forms of the anisotropic system of equations based on the effective tensorial properties of the elliptical inclusions at various angular orientation. Here, we numerically investigate the macroscopic models in order to analyze fingering behavior resulting from the effective diffusion tensors. Our method is based on the finite elements on a rectangular domain covered with a triangular mesh. The values of the physical quantities used in the numerical simulation is given in Table 1. Fig. 8(a) shows the evolution of the longitudinal, \( \psi_{L} \), and transverse, \( \psi_{T} \), Lewis numbers with \( \theta \). The monotonic nature of the Lewis numbers as a function of \( \theta \) suggests that the pattern behavior for each value of \( \theta \) is different. Moreover, the behavior of the patterns at \( \theta = 0^\circ \) and \( \theta = 90^\circ \) predicts respectively two distinct states; sparse pattern when \( \sigma \ll 1 \) and cusp-like (connected) pattern when \( \sigma \gg 1 \). Alternatively, we can also say that, at the two extreme values of \( \theta \), the system is dominated by mass diffusion (\( \psi_{T} \to 0 \)) and heat diffusion (\( \psi_{T} \to \infty \)). It should be noted that the values of \( \psi_{L} \), \( i = (L, T) \), at the right half of the range of values of \( \theta \) has similar monotonic behavior, but in the reverse directions. In Fig. 8(b), the evolution of the anisotropy indicators is depicted.
At $h = 0$, $90^\circ$, $180^\circ$, the system is diagonally anisotropic with $\mu = 0$. As $h$ increases away from $0^\circ$, the effect of anisotropy becomes more pronounced in the system, which is mainly characterized as symmetrically anisotropic. The anisotropy effect gives rise to distinct nonuniformities on the fingers. At $h = 0^\circ$, $180^\circ$, the patterns are qualitatively alike. Fig. 9 depicts the behavior of char patterns at various values of $h$. The patterns corresponding to the set of values of $h = (15, 165)\ldots,(75, 105)\ldots$ are reflected and qualitatively similar, since the orientation of $h$ changes the sign of the off-diagonal entries of the effective tensors. In Fig. 9(a) and (b), it is clear that the patterns do not form clusters, which is a characteristic feature of diagonally anisotropic tensors. Fig. 10 depicts reflected char patterns at different values of $h$. The pattern behavior is reflected mostly for $h = 105^\circ$, $120^\circ\ldots$, since $\Phi$ changes sign. Thus, these values of $\theta$ can be regarded as reflections of $h = 75^\circ$, $60^\circ\ldots$, $15^\circ$. Interestingly, in Fig. 10(c) and (d), the patterns are qualitatively similar, but the reflection at $h = 135^\circ$ indicates enhanced clustering of the fingers. The clustering of fingers leads to higher tendency for screening effect on adjacent fingers, which suppresses their growth [1]. The enhanced pattern behavior for reflection angles is also true for other values of $h$ on the right half range, except for $h = 180^\circ$. Further, the nonuniformity on the patterns increases from $h = 30^\circ$ to $h = 45^\circ$ and then gradually decreases in intensity as the indicators ($\gamma; \mu$) decreases toward $h = 90^\circ$ (see Fig. 8(b)), in which case the patterns are more cusp-like and connected (see Fig. 8(b), and Fig. 11(a) and (b)). Fig. 11(a) and (b) shows reflected patterns for $h = 75^\circ$, $105^\circ$. The nonuniformity on the patterns decreases as $\gamma$ decreases. The patterns are more connected, which indicates the onset of the formation of cusp-like patterns. We also compared the pattern behavior with the isotropic case in which $\gamma = 1$ and $\mu = 0$. It should be noted that the effective diffusion constants for the numerical simulation depicted in Fig. 11(d) are calculated on an isotropic geometry with the same volume fraction as those for the anisotropic case. It is clear that the pattern is not directed toward the lateral boundary, but propagates along the direction of oxidizer flow.

5.2. Fingering behavior based on thermal-diffusion instability

The approach of the previous subsection is strictly based on the effective anisotropic diffusion tensors computed through the homogenization method. For that purpose, our goal was to discuss the relationship between the local structures and the macroscopic fingering behavior for general anisotropic media. In this sense, we are not particularly focused on the distinct fingering regime. In this subsection, however, we illustrate the anisotropy effect on the fingering patterns for the distinct regimes. Thus, we approach the problem from a viewpoint of the mechanism of thermal-diffusion instability [see 34–37, e.g.]. The objective is to show how
Fig. 12. Spatial profiles of char patterns for a generic diagonally anisotropic material. All regimes of the pattern behavior can be reproduced. Fingering behavior at (a) \( \gamma = 0.35 \); (b) \( \gamma = 0.6 \); (c) \( \gamma = 0.8 \); (d) \( \gamma = 1.0 \) (isotropic case); (e) \( \gamma = 1.2 \); (f) \( \gamma = 1.4 \). The numerical simulations correspond to the tip-splitting regime at \( \text{Le} = 0.1 \); \( \text{Pe} = 10 \).

Fig. 13. Spatial profiles of char patterns for a generic diagonally anisotropic material. In the computations \( \gamma = 0.5 \); \( \mu = 0 \). From (b)–(f), \( \text{Pe} = 10 \). (a) \( \text{Le} = 0.015 \); \( \text{Pe} = 0.1 \) (sparse fingering regime); (b) \( \text{Le} = 0.1 \) (tip-splitting regime); (c) \( \text{Le} = 0.2 \); (d) \( \text{Le} = 0.5 \); (e) \( \text{Le} = 0.7 \); (f) \( \text{Le} = 1 \) (regime of connected front). The distinct fingering regimes can be reproduced and nonuniformity on the patterns is not pronounced.
anisotropy affects the uniformity as well as reproducibility of the distinct regimes of the fingering patterns. Here, the boundary condition for the heat equation is modified by prescribing the following external ignition source at the left boundary to mimic the onset of ignition

\[ T_{\text{ign}}(t) = \begin{cases} T_1, & \text{if } 0 < t < t_1 \\ T_0, & \text{otherwise.} \end{cases} \]  

(35)

where \( T_0 \) and \( T_1 \) are constants. As before, we prescribe a Dirichlet boundary condition, i.e. \( T = T_0 \) at the right boundary.

5.3. Fingering behavior in diagonally anisotropic medium

The mechanism of thermal-diffusion instability can now be used to analyze pattern behavior for a “weakly” anisotropic material by considering a generic diagonally anisotropic medium satisfying \( \mu = 0 \) and \( 0 < \gamma < 2 \) in (31). The pattern behavior is said to be weak if the fingering patterns are not strongly deviated from the direction of flow and nonuniformity is not characterized by finger-like clusters. The problem in an isotropic medium can immediately be recovered from this system by setting \( \gamma = 1 \). Fig. 12 shows the spatial profiles of char patterns as a function of the anisotropy indicator \( \gamma \). The fingering patterns are considered in the tip-splitting regime, which corresponds to \( Le = 0.1 \) and \( Pe = 10 \). The numerical results discussed here are for a weakly anisotropic medium, in which the uniformity and reproducibility of the fingering behavior is not significantly affected. The anisotropy is such that the population (density) of the fingers is changed. For values of \( \gamma < 1 \), the fingering patterns are dense (see Fig. 12(a)-(c)). However, for some values of \( \gamma > 1 \), the fingers become more distinct from each other (see Fig. 12(e) and (f)). This characteristic behavior accounts for the effect of transverse diffusion that manifests at \( \psi^{-1}_t > \psi^{-1}_l \), which increases the spacing between adjacent fingers. Fig. 12(d) corresponds to the isotropic case at \( \gamma = 1 \), which has been included simply for comparison. It can be seen that pattern behavior for diagonally anisotropic media have qualitative resemblance to those of isotropic media, except for the increase in the density of fingers. In Fig. 13, the spatial profiles of char patterns at distinct fingering regimes are illustrated. The numerical simulations were calculated for an anisotropy indicator, \( \gamma = 0.5 \). The regime of sparse fingers is depicted in Fig. 13(a). The pattern behavior is qualitatively similar to the isotropic case since the nonuniformity of the pattern is not obvious. Fig. 13(b) shows the tip-splitting regime. By increasing the Lewis number from \( Le = 0.2 \) to \( Le = 1 \), we arrive at the regime of connected front. (see Fig. 13(c)-(f)). Thus, we conclude that the distinct fingering regimes can be reproduced for problems posed in a diagonally anisotropic medium. In the computations, the anisotropy indicators are set to \( \gamma = 0.5 \), and \( \mu = 0 \).

5.4. Fingering behavior in a symmetrically anisotropic medium

For the considered problem in a medium with elliptical inclusions and having distinct values of \( \theta \), the majority of the tensorial realizations of the effective diffusion quantities can be characterized as symmetrically anisotropic. For example, for values of \( \theta = 30^\circ, 45^\circ, 60^\circ \). Here, we consider anisotropic media with elliptical inclusions at \( \theta = 45^\circ, 135^\circ \) as instances of a symmetrically anisotropic medium. In Fig. 14, the spatial profiles of distinct fingering regimes of the char patterns are illustrated for the problem posed in a symmetrically anisotropic material, in which the inclusions are oriented at \( \theta = 45^\circ \). In this case, the material is highly anisotropic, since nonuniformity on the patterns is pronounced. In all the computations, \( \gamma = 1 \), \( \mu = 0.2 \). Fig. 14(a) corresponds to
In Fig. 15 (a)–(d), diffusivity tensors calculated by the homogenization method. The medium consisting of periodically distributed elliptical inclusions is considered by investigating the behavior of the patterns on a porous medium. The latter has been manifested in all the fingering regimes, the distinct fingering states are still reproducible in the anisotropic medium. This confirms that material anisotropy is not significant for the emergence of the distinct fingering states. In Fig. 15, we illustrate the spatial profiles of char patterns for a problem posed in a symmetrically anisotropic material with inclusions at the spatial profiles of char patterns for a generic symmetrically anisotropic material with $\theta = 135^\circ$. From (a)–(d), $\gamma = 1$, $\mu = -0.085$, $-0.15$, $-0.2$, $-0.25$. The fingers are dense; (f) for $\gamma = 1.5$, $\mu = -0.2$, the patterns are more distinct due to increase in $\gamma$.

The regime of sparse fingers at $Le = 0.02$. It can be seen clearly that the uniformity of the fingering patterns is reduced, since the fingers are directed toward a lateral boundary. The tip-splitting regime is showed in Fig. 14(b). In this case, the nonuniformity on the pattern is also obvious. Similarly, for increasing values of the Lewis number, i.e. from $Le = 0.25$ to $Le = 1$, we approach the regime of connected front (Fig. 14(f)). While the fingering behavior manifested nonuniformity in all the fingering regimes, the distinct fingering states are still reproducible in the anisotropic medium. This confirms that material anisotropy is not significant for the emergence of the distinct fingering states. In Fig. 15, we illustrate the spatial profiles of char patterns for a problem posed in a symmetrically anisotropic material with inclusions at $\theta = 135^\circ$. In this case, the anisotropy indicator $\mu$ changes sign, which reflects the patterns. The patterns are directed toward the right lateral boundary compared to the fingering behavior depicted in Fig. 14. In Fig. 15(a)–(d), $\gamma = 1$, $\mu = -0.085$, $-0.15$, $-0.2$, $-0.25$. The nonuniformity on the patterns increases for increasing values of $\mu$. In Fig. 15(d), the nonuniformity on the pattern is extended to the right lateral boundary, on which the fingers propagate. Fig. 15(e) corresponds to the case $\gamma = 0.5$, $\mu = -0.2$; the pattern show increased density of fingers and propagation on lateral boundary. In Fig. 15(f), the fingering patterns are distinct due to the increase in transverse diffusion parameter at $\gamma = 1.5$.

6. Conclusion

In the present paper, we analyzed the behavior of fingering char patterns in terms of material anisotropy. The latter has been considered by investigating the behavior of the patterns on a porous medium consisting of periodically distributed elliptical inclusions. Specifically, anisotropy was accounted for in the system of balance equations through the effective thermal conductivity and mass diffusivity tensors calculated by the homogenization method. The numerical values of the tensors were calculated for different angular orientations of the elliptical inclusion of the REV. The tensors revealed distinct tensorial properties such as diagonally anisotropic and symmetrically anisotropic. Thus, for each angular orientation, numerical simulation was performed on the corresponding macroscopic medium with constant diffusion tensors. Our results exhibited two distinct limit behaviors corresponding to cases in which the tensors are diagonally anisotropic. First, at $\theta = 0^\circ$, the numerical result is reminiscent of the sparse finger-like pattern, which manifest in a diffusion-dominated system. The behavior of the fingering pattern at the diffusion-dominated regime is consistent with the behavior observed in the literature [see 10,12,38, e.g.], and in other context [see 39,40, e.g.] for a non-adiabatic situation. Thus, we conclude that the transverse diffusion in the principal direction is significant for the emergence of the finger-like patterns in diffusion-dominated systems. In the second case at $\theta = 90^\circ$, the pattern is cusp-like and corresponds to the limit of large flows. However, for all values of $\theta$ between the limits, the patterns are nonuniform with a peak anisotropy effect at $\theta = 45^\circ$, respectively $\theta = 135^\circ$ in which the finger-like patterns are in the form of clusters. The numerical results showed that a diagonally anisotropic system has less observable anisotropy effect on the fingering patterns. In terms of directional fingering, the morphology of the patterns for such systems is almost identical to the isotropic case, except for the density of fingers that changes with the anisotropy indicator.

It should be pointed out that an observable nonuniformity effect, in the classes of anisotropic systems considered, has been described as the effect that tends to deviate the fingering patterns from the principal direction of flow, promote the formation of clusters, and in some cases the propagation of the pattern on the lateral boundary. Our strategy based on the homogenization method on fixed periodic structures reproduces some forms of nonuniform pattern behavior such as clustering of fingers, directional fingering...
and propagation on the lateral boundary. Having said that, the different pattern behaviors and realizations based on the angular orientations of the elliptical inclusion may be taken to be local within the porous material of interest and hence collectively they showed that the global pattern behavior is indeed nonuniform in the whole sample. In the next step, we examined the pattern behavior using the mechanism of thermal-diffusion instability. We conclude that for a highly anisotropic (i.e. symmetrical anisotropy in the considered problem) media, the uniformity of the fingering behavior is reduced, otherwise the distinct experimentally observed fingering regimes manifested, i.e. material anisotropy has no significant effect on the fingering behavior. This observation is consistent with the experimental work of [1,6,7].

Moreover, we also mention that the homogenization ansatz (15) used for the derivation of the upscaled problems [21]–[24] fails to work in the presence of sharp gradients at the level of the REV. However, the exposition of the present paper is sufficient at the macroscopic level, where the presence of sharp gradients is indeed of little consequence. Thus, in view of the relationship between the solution to the problem at the REV and the results presented in this work, we expect, in spite of the difficulties inherent in the microscopic point of view, that the results described in this work closely approximate the microscopic problem. However, in the presence of sharp gradients as evident in the macroscopic results, a possible test of the relationship, bearing in mind the method of asymptotic homogenization, would be to work in line of argument recently introduced in [41], in which the authors described a new methodology to deal with cases where the classical homogenization theory breaks down, e.g. the presence of sharp fronts. In this direction, a similar averaging approach, [42], based on extension to the volume averaging method described in [43,44], can also be adopted. In an upcoming review of the present work, we analyze a modified premise of our homogenization approach through the two-scale asymptotic expansion method with drift [41].

Furthermore, still open in this research area are a detailed linear stability analysis for highly anisotropic media and the influence of other filtration properties of the porous media. The latter might potentially lead to other aspects of the problem, specifically the hydrodynamic behavior of patterns, which have been reported in other contexts [see 45–49, e.g.]. In the present study, the flow field has been considered to be uniform. It might also be of research interest to understand the mechanism of the propagating char patterns in a nonuniform flow environment.

Conflict of interest

None declared.

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References


