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## Precipitation dendrites in channel flow

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**Abstract** – Precipitation on rough walls under a channel flow of a supersaturated fluid can lead to dendrite pattern formation. A passive advection of a solute has a great influence on the dendrite morphology similar to the convection effect on the solidification dendrites emerging from an undercooled liquid. We show how the asymmetric dendrite growth depends on the solute advection and turbulent mixing. Implications of our study on the generic mineral scale formation phenomena are also discussed.

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**Introduction.** – The growth of solid precipitating structures, also known as scale formation, is a common and costly phenomenon in many industrial processes that deal with water or other fluid-handling systems (*i.e.* wells, heat exchangers, tanks and delivery lines, etc.), where mineral precipitation in pipes, on equipment or as fracture filling has a detrimental effect on process efficiency, cost and lifetime of processing technologies. Scale formation is encountered in a large number of industries including paper-making, chemical manufacturing, cement operations, food processing, as well as non-renewable, (*i.e.*, oil and gas [1]) and renewable (*i.e.*, geothermal, see fig. 1) energy production and medical instruments.

Albeit precipitation patterns have been theoretically studied for many decades, most of the matter-of-fact concerns regarding the controlling factors that influence scale formation or inhibition in pipelines, and the effect this may have on the coexisting corrosion processes are still open to investigations. In particular, many of these pipelines can sustain turbulent flows and, in this regime, the effect of turbulent mixing on mineral scale formation is not yet fully explored and understood.

Interestingly, many of the precipitation patterns are rather robust with respect to changes in the detailed aqueous chemistry and interfacial composition, and often similar to the surface patterns developed during freezing from a supercooled liquid [2,3]. The similarity between different freezing and precipitation morphologies suggests a deeper connection between these deposition mechanisms,

and the possibility of modelling deposition pattern formation within a generic theoretical framework. In fact, there are several approaches to modelling the dynamics of liquid-solid interfaces, among the most common ones being based on boundary tracking methods, such as the level set method, *e.g.* [4] and the phase-field method, *e.g.* [5].

Numerous studies document that solidification patterns using phase-field modelling [5,6] have similar properties as those observed in nature and experiments [7,8]. Recently, a phase-field model for precipitation has been proposed as an efficient tool to study surface morphology developed by precipitation from a supersaturated solution [9,10]. For certain conditions of supersaturation and interfacial energy, the precipitation surface develops dendritic structures similar to those developed during solidification from an undercooled liquid in hydrostatic equilibrium.

Motivated by the problem of mineral scaling in the presence of turbulent mixing, we study the precipitation dendrite morphologies under different hydrodynamic conditions using a phase-field approach combined with the lattice Boltzmann method. Albeit, the effect of laminar flow on solidification dendrites is well studied [11–13], the non-laminar regime that allows for turbulent mixing downstream of the crystal growth remains virtually uncharted. The shear flow has a great influence on the rate of growth of the crystal, as well as on the orientation of the dendrites with respect to the main flow direction, and the spacing between dendritic branches. Also, the lateral growth of dendrites is enhanced in the upstream direction,



Fig. 1: (Color online) Amorphous silica  $(SiO_2)$  mineral precipitation from geothermal waters in a retention tank at the Nesjavellir Power Plant, Iceland (from ref. [14]). Bulk flow velocity is 0.03–0.06 cm/s depending on operating conditions.

and suppressed in the downstream when the flow remains relatively smooth. Our aim is to investigate the effect of non-laminar downstream flow on the dendrite morphology.

The rest of the paper is organized as follows: In the second section, we introduce a two-dimensional (2D) model of crystal growth nucleated on one of the pipe walls under laminar and turbulent flow. We continue in the third section with a numerical implementation of this model based on a coupling between a phase-field method for surface precipitation and an augmented lattice Boltzmann method for turbulent flows. The fourth section presents the numerical results on the precipitation dendrites and the influence of solute advection and turbulent eddies on the growth rate and dendrite morphology. Finally, we offer a discussion and concluding remarks in the fifth section.

**Precipitation under flow.** – We study the coupling of surface precipitation with fluid flow in a 2D setup corresponding to a longitudinal cross-section of a pipe of diameter H and length L. For simplicity, we consider that the pipe is horizontal and the effect of gravity is neglected, *i.e.* the regime of precipitation growth is isolated from that of sedimentation of the solute particles. On one of the walls, there is an initial asperity represented by a half circle of small radius and located in the middle of the pipe length.

A supersaturated fluid is injected at the pipe inlet and removed at the outlet. The soluble particles are described by a concentration field c(x, y, t) that satisfies the diffusion-advection equation

$$\frac{\partial c}{\partial t} + \mathbf{v}_f \cdot \nabla c = D \nabla^2 c, \tag{1}$$

where D is the solute diffusivity coefficient and  $\mathbf{v}_f$  is the fluid velocity. We assume generic first-order kinetics at the wall side with an asperity located at  $(x, y) = \Gamma_t$ , where  $\Gamma_t$  is a parametrization of the precipitation surface. Thus, the boundary condition at the reactive site is

$$D\nabla c \cdot \mathbf{n} = k(c - c_{eq}), \quad \text{on } \Gamma_t, \tag{2}$$

since  $\mathbf{v} \cdot \mathbf{n} = 0$  at  $\Gamma_t$  meaning that the crystal is impermeable to the moving fluid. Here, k is a constant reaction rate,  $c_{eq}$  is the equilibrium concentration, and  $\mathbf{n}$  is the surface normal unit vector pointing towards the fluid phase. We assume that the walls are inert and rigid, *i.e.* no solute diffusion or deformation inside the walls. Equation (2) expresses that the mass transport to the surface equals the rate of precipitation. By this process, the interface advances into the fluid with a normal velocity  $(v_n)$  that is given by

$$\rho_s v_n = k \rho_f (c - c_{eq}), \quad \text{on } \Gamma_t, \tag{3}$$

with  $\rho_s$  being the mass density of the solid. The pipe flow of an incompressible fluid is described by the Navier-Stokes equation

$$\frac{\partial \mathbf{v}_f}{\partial t} + \mathbf{v}_f \cdot \nabla \mathbf{v}_f = -\frac{1}{\rho_f} \nabla p + \nu \nabla^2 \mathbf{v}_f, \qquad (4)$$

where p(x, y, t) is the pressure field,  $\nu$  is the uniform kinematic viscosity, and  $\rho_f < \rho_s$  is the constant fluid density which results, by the conservation of mass, in a solenoidal velocity field condition  $\nabla \cdot \mathbf{v}_f = 0$ .

We use zero-flux boundary conditions on the lateral walls, y = 0 and H, for the pressure and concentration fields, and no-slip boundary conditions for the velocity field. The concentration field is constrained by the surface growth conditions given by eqs. (2), (3) and applied on  $\Gamma_t$ . At the inlet x = 0, the injected fluid has a fixed Poiseuille velocity profile and a given supersaturation level of the concentration field  $c > c_{eq}$ . Initially, the concentration is uniform and equal to equilibrium value everywhere else away from the pipe inlet. The supersaturated fluid is then removed at the outlet by setting open-flux boundary conditions at x = L.

Using a typical lengthscale  $x_0$ , timescale  $t_0$  and equilibrium concentration  $c_{eq}$ , all the dimensional variables can be converted into dimensionless forms as  $\tilde{t} \equiv t/t_0$ ,  $\tilde{x} \equiv x/x_0$ ,  $\tilde{c} \equiv (c - c_{eq})/c_{eq}$ ,  $\tilde{p} \equiv p/p_0$ ,  $\tilde{\mathbf{v}} \equiv \mathbf{v}/v_0$ , and  $\tilde{v}_n \equiv$  $v_n/v_0$ , where  $v_0 = x_0/t_0$  and  $p_0 = \rho_f x_0^2/t_0^2$ . By inserting the rescaled variables into eqs. (1)–(4) and dropping the tildes, the following set of evolution equations is obtained:

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = \frac{1}{Pe} \nabla^2 c,$$
  
$$\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} = -\nabla p + \frac{1}{Re} \nabla^2 \mathbf{v},$$
 (5)

which is complemented by the aforementioned boundary conditions and the rescaled interfacial conditions given as

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$$\mathbf{n} \cdot \nabla c = Da \cdot c,$$
  
$$v_n = \beta \frac{Da}{Pe} c, \quad \text{on } \Gamma_t,$$
 (6)

where  $\beta = c_{eq}\rho_f/\rho_s$ . The group of dimensionless parameters governing the transport and precipitation kinetics is given by the Péclet number  $Pe = v_0 x_0/D$ , that is the ratio of the rate of fluid advection to the rate of solute diffusion, the Reynolds number  $Re = x_0v_0/\nu$ , that measures the fluid advection relative to viscous transport, and the Damköhler number  $Da = kx_0/D$ , that measures precipitation rate relative to the diffusion transport rate.

A typical lengthscale that remains constant is, for instance, the diameter of the pipe, so we can set  $x_0 \equiv$ H. A typical flow velocity can be set by the injection velocity  $v_0 \equiv V$ . Also  $\rho_f / \rho_s \approx 1/2$ , but the equilibrium concentration (mass fraction) is typically a small number  $c_{eq} \sim \mathcal{O}(10^{-3})$ . So,  $\beta$  would be, in general, a small number. In the regime where  $Da \ll Pe$ , we can safely assume that  $v_n \approx 0$ , given that also  $\beta \ll 1$ . For surface growth, we are interested in the opposite limit corresponding to  $Da \gg Pe$ , which is equivalent to  $v_0 \ll k$ , or that the typical flow velocity is much smaller than the kinetics rate (notice that the interfacial kinetics rate has units of [k] = m/s as velocity). From this point of view, fast-growing dendrites are observed when the precipitation growth happens on a much faster timescale than the typical fluid flow. This allows the underlying effects of advection and *Re* number on the morphology to be studied on much shorter simulated timescales.

Phase field and lattice Boltzmann. – We use a phase-field method to efficiently track the precipitation front. The phase-field modelling has been extensively used as a powerful tool to handle the evolution of complex microstructures. Various thermodynamically consistent formulations have been proposed for the dendritic solidification problems, but they have similar numerical properties [5,15]. Recently, a phase-field formulation of a precipitation front has been proposed in refs. [9,10] in the absence of fluid flow. Here we extend the model to also include this effect. The evolving precipitation surface is tracked as the zero level of an auxiliary phase field  $\phi(x, y, t)$  which varies rapidly across the liquid-solid interface, and takes homogeneous values in the bulk phase,  $\phi = -1$  in the solid and  $\phi = 1$  in the fluid. In the dimensionless form, the governing equations for the phase and concentration fields read as

$$\frac{\partial \phi}{\partial t} = \frac{P_{er}}{P_e} \left( \nabla^2 \phi - \kappa |\nabla \phi| \right) - \frac{\partial f}{\partial \phi},$$

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = \frac{1}{P_e} \nabla^2 c + \frac{1}{2\beta} \frac{\partial \phi}{\partial t}$$

$$+ \frac{P_e}{2\beta D_a} \frac{\partial \phi / \partial t}{|\nabla \phi|} \left( \frac{1}{P_e} \nabla^2 \phi - \frac{\partial \phi}{\partial t} \right), \quad (7)$$

where  $P_{er}^{-1} = D\tau/\epsilon^2$  is the interfacial Pe number, defined in units of the interface thickness  $\epsilon$  and phase-field characteristic timescale  $\tau$ ,  $\kappa = \nabla \cdot (\nabla \phi/|\nabla \phi|)$  is the local curvature, and  $f(\phi, c) = \lambda c(\phi - \phi^3/3) - \phi^2/2 + \phi^4/4$  is the local free energy that has two minima corresponding to the liquid and solid phases. Equation (7) includes two additional terms that implicitly represent the interfacial boundary conditions. The first term acts as a net source or sink of solute due to the discontinuity in the solute

concentration gradient across the interface, whereas the second term corresponds to a net flux of solute arising from the discontinuity in the solute concentration across the interface. As shown in ref. [9], the phase-field formulation in eqs. (7) converges in the limit of  $P_{er} \rightarrow 0$  to the sharp-interface equations (5), (6). These partial differential equations are solved on a regular grid using an isotropic finite difference method where both nearest- and next-nearest-neighbouring grid points are considered.

The pipe flow is numerically simulated using the lattice-Boltzmann-method-type LBGK [16] with 9 principal directions in 2 dimensions (D2Q9). This is augmented with a subgrid model to effectively resolve the turbulent energy transport at high *Re* numbers at lengthscales below the grid resolution. In this case we implement the Smagorinsky subgrid model [17,18] which modifies the local viscosity  $\nu_{eff}$  to include a turbulent or eddy viscosity  $\nu_{eff} = \nu + \nu_t$ ,  $\nu$  being the kinematic viscosity the eddy viscosity  $\nu_t$  is calculated from the filtered rate of strain tensor  $\dot{\epsilon}_{i,j} = (\partial_j v_i + \partial_i v_j)/2$  as  $\nu_t = (C_s \Delta)^2 \dot{\epsilon}$ , where  $\dot{\epsilon} = \sqrt{2 \sum_{i,j} \dot{\epsilon}_{ij} \dot{\epsilon}_{ij}}$ ,  $\Delta$  is the filter width, and  $C_s$  is the Smagorinsky constant.

To implement no-slip conditions at the solid-liquid interface we use bounce-back boundaries in the lattice Boltzmann model. The position of the interface within the lattice Boltzmann is constantly updated by setting all cells with  $\phi < 0$  as solid which adhere to the bounceback rules. Within the liquid part of the moving interface the velocity is dissipated by increasing the viscosity, *i.e.*  $\nu = \nu_0 2/(1 + \phi)$ , as  $\phi \to 0$  [19].

The implementation of the lattice Boltzmann model is benchmarked against known solutions for pressure-driven channel flow for low and high Re numbers, the error corresponds to that obtained for by other implementations of the same type [20]. Further tests regarding the generation of turbulent structures shows qualitative agreement when compared against results generated for flow past including objects [16].

**Results and discussion.** – In order to simulate the effect of diffusion, advection and turbulence upon the morphology of precipitating structures we perform a number of simulations for various Pe and Re numbers.

In fig. 2, we depict the effect that fluid transport has on the dendrite morphology. The fluid streamlines and the concentration profile away from the precipitation dendrite for different Pe and Re numbers are shown in fig. 3. The precipitation area that protrudes furthest into the high flow region experiences the highest advection rates, and hence the steepest concentration gradients. Downstream of this area, a separation of flow rates, caused by the barrier which the growth presents to the fluid, creates a region of low fluid velocity where advection is negligible. Figure 4 shows the evolution of the growing structure, and the clear asymmetry in the growth as advection takes over the diffusive transport of the solute. The average



Fig. 2: (Color online) A sketch of the setup and the anisotropic dendrite growth due to fluid advection.



Fig. 3: (Color online) Normalized concentration field with the fluid streamlines superimposed. Inside the crystal the rescaled concentration relaxes to the equilibrium value, whereas far away from the interface it reaches a constant supersaturation level in the fluid.



Fig. 4: (Color online) Evolution of the precipitated growth for various Pe and Re numbers. Contour lines show the position of the precipitation front at different times.



Fig. 5: (Color online) The angle of growth plotted at a fixed time for different values of Pe and Re numbers. The curves trace the numerical data points using a spline interpolant.

asymmetric growth angle about the location of the initial rough element is calculated in fig. 5. The nature of the curve is a result of the competition between different transport timescales. The initial symmetry at low Penumbers is related to the diffusion-dominated growth. The advective transport increases the growth rate in the region it affects, hence maximum asymmetry occurs when advection affects only the main dendrite tip (Pe = 1-5). At higher advection rates (Pe > 5) a larger area is affected and symmetry increases. Further discussion will therefore refer to the effects various flow regimes have on this asymmetry, and this can be divided into several key segments (see fig. 2 for reference).

Diffusion-dominated Pe < 1. The precipitation reaction removes the dissolved precipitate from the fluid producing a solute-depleted region near the interface. Advection and diffusion are the main transport processes by which the depleted concentration is advected or redistributed. For Pe < 1, advection has a negligible effect, as the system is dominated by diffusive transport that leads to a symmetric redistribution of the solute depletion, and thus the growth is also axial symmetric.

Transition region Pe = 1-5. The transition region between the diffusion-dominated and advection-dominated regimes produces most asymmetric dendritic growths. With increasing Pe number, the advection becomes dominant especially at the furtherest upstream tips of the precipitation dendrites, where the lower supersaturation is advected downstream, and, henceforth, replaced by higher levels of supersaturation that promotes faster growth. Consequently, the areas immediately downstream of this are either supplied with less supersaturated fluid, or lower flow rates behind an obstacle and diffusion-dominated growth. In both cases, the precipitation surface is restricted to grow slower downstream then upstream, which produces the observed asymmetry. Advection-dominated Pe > 5. Higher rates of advection expose a greater section of the surface to supersaturated fluid, while the more solute-depleted fluid is advected downstream. This effectively increases the areas of faster growth into the downstream direction, thereby the growth asymmetry is overall reduced. In the asymptotic limit of  $Pe \rightarrow \infty$ , the flow rate becomes so high that the depleted supersaturation is "instantly" advected away from the precipitation site. At low Re numbers, the standing vortex formed will still force an area of the solid surface to exist in a diffusion-dominated regime, hence we suspect that the crystal growth will never become fully symmetric. This is indicated by the curves in fig. 5 for lower Renumbers, where the curves appear to saturate at high Pe.

Effect of Re number. The effects caused by increasing *Re* number are two-fold. Firstly, the precipitation dendrite represents an obstacle in the fluid flow, causing lower flow velocities and a large standing eddy to form downstream behind the obstacle (see the streamlines in fig. 3). With increasing Re number, the size of this downstream region expands, and isolates a greater region of the solid surface. With little advection the growth is diffusion dominated and slow relative to advected areas. Increasing *Re* number therefore promotes asymmetry as it forces a larger quantity of the surface downstream to grow less rapidly. The second impact is observed within the velocity field as the fluid approaches the solid boundary. At higher Re numbers, the gradients of the velocity field are much sharper at the boundary due to no-slip boundary conditions. This forces a greater portion of the solid surface to experience high advection rates, specifically in the areas further upstream of the fastest-growing point. In turn, this pushes a greater quantity of depleted fluid downstream further reducing the growth rates in the area immediately adjacent to the fastest-growing regions, see fig. 4.

Turbulence. Turbulent eddies are generated by the induced surface dendrites generated from precipitation. At high *Re* numbers, the stable vortex, that forms downstream and behind the dendrite, evolves into a turbulent puff where eddies shed from the grown structure and propagate downstream. Thus, the precipitation of the downstream side of the dendrite is directly affected by the concentration mixing inside the turbulent puff. Turbulent mixing promotes effectively an isotropic transport of supersaturation, and, in this regime, the advective transport can be modelled as an effective isotropic diffusive process with an enhanced, effective solute diffusivity that depends on the flow pattern [21].

The enhanced turbulent diffusivity and isotropic distribution of concentration gradients accelerates the growth rate on the downstream side, and reduce the overall asymmetry of the pattern. This can be clearly seen in fig. 4, where dendrites on the right-hand side are more prominent for high Re numbers. The size of the area affected by turbulent mixing is dependent upon the orientation angle at which the main dendrite (which blocks



Fig. 6: (Color online) Normalized concentration field for precipitation growth upon a pipe wall with randomly distributed rough elements. Pe = 10, Re = 10000 (top), Re = 100(bottom). The superimposed black lines correspond to fluid streamlines (flow from left to right), whereas the white lines represent the time evolution of the precipitates by showing the position of the precipitation surface at different times.

the flow) grows relative to direction normal to the flow. At shallow orientation angles, a greater region downstream of the blocking dendrite experiences the enhanced diffusivity due to turbulent mixing. Figure 5 shows that for a small window of shallow growth angles, turbulence causes the curve for Re = 10000 to become more symmetric than the curve for Re = 5000. With increasing Pe number, the turbulent puff promotes more downstream mixing to occur on a timescale considerably shorter then the growth rate. It may be assumed that the growth would therefore once again become symmetric as  $Pe \rightarrow \infty$ . However, since the mixing in the turbulent puff depends upon a shallow growth angle, a symmetric state may never be reached. Here, the high growth angles prevent the turbulent eddies from altering the concentration gradients in every direction, and asymmetry is therefore maintained.

Rough interface. – Precipitation in pipe flows can nucleate at any asperity on the pipe walls, and the interaction between different growing sites through the flow field around them can generate new effects and morphologies that are not captured by the dynamics of an isolated precipitate. Henceforth, it is interesting to extend our model to also include the precipitation on a rough interface. In fig. 6, we show few preliminary results on the morphology of an evolving rough interface that initially consisted of 14 asperities of an equal size, and that are randomly and uniformly distributed along one of the walls. We notice that the non-laminar downstream flow has a drastic effect on downstream precipitates. More compact, columnar structures are promoted with increasing Renumber due to turbulent mixing. We also observe longrange interactions mediated by the surrounding flow field and a "screening" effect that the precipitates induce on each other. A detailed analysis of the correlations between evolving precipitates in turbulent pipes and a comparison with the precipitates formed in hydrothermal pipelines (fig. 1) constitute however the subject of another report.

**Conclusion.** – Precipitation upon a single rough element within a pipe has been simulated at high advection rates and turbulent Re numbers by coupling a phase-field

method for precipitation and a lattice Boltzmann method for fluid flow. We have shown that the morphology and growth orientation of the precipitate is greatly influenced by the flow conditions. At low advection rates, where diffusion controls the system, the precipitation dendrites grow symmetrically. As the advection increases relative to diffusion, the system becomes rapidly asymmetric since a small portion of the solid surface becomes exposed to supersaturated fluid. This promotes the precipitate to grow upstream relative to the flow direction. Maximum asymmetry and shallowest normal growth occur in the region where advection overtakes diffusion as the dominant transport mechanism. Higher advective transport reduces the growth asymmetry due to the efficient transport of the depleted solute. Furthermore, with increasing Reynolds number the growth asymmetry also increases, however, the turbulent mixing at high Re numbers leads to a more symmetric growth.

Our results also show that the fast precipitation dominated by advection leads to more compact growth structures that tend to develop less dendritic branching, as seen from figs. 3 and 4. This compact growth occurs over a larger area for higher Re numbers where steep velocity gradients expose more of the solid interface to high advection rates. Turbulent mixing away from a precipitation site has a major effect on the columnar growth of the downstream precipitates.

Mineral scale formation often leads to the generation of rough internal pipeline surfaces. It occurs in a wide variety of systems at a wide range of fluid flow rates and Reynolds numbers and the growth of rough walls further complicates the flow dynamics. This is the first attempt to couple mineral precipitation and hydrodynamics for pipeline flow rates above the strictly laminar regime.

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