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Impact of hydrodynamics on growth and morphology of faceted crystals

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ABSTRACT

The growth of faceted crystals occurs often in nature and industry, involving often the presence of flow. The growth of faceted crystals is the result of interface kinetics and diffusion phenomenon. The present paper presents a front tracking interface model based on a cellular automaton approach for the simulation of faceted crystal growth. The current model takes into account the interface kinetics and solute transport by diffusion and convection. The propagation of kinks is modelled by differentiating two growth velocities, one normal and one lateral at each face. The positions of the crystal corners are shifted according to growth of adjacent faces. The hydrodynamics is computed with a two-phase model using a penalty method to model the presence of growing obstacles (the crystals). This model was applied in 2D to the growth of hexagonal Fe₂Al₅ crystals, so called top dross particles, in a saturated liquid at constant temperature. Qualitative comparison was made between simulation and experimental observation of crystal shape and size. The growth rate was found to be strongly influenced by the flow hydrodynamic induced kinetics.

1. Introduction

Computational models for solidification take into account two physical phenomena: 1) the attachment kinetics that describes the molecular dynamics at the solid interface, and 2) the particles transport via diffusion towards the growing interface. Some diffusion-limited models are explicitly tracking the interface in a Lagrangian way, they are known as sharp front-tracking models. The liquid-solid interface is localised only inside one volume element. Other diffusion-limited models such as the phase-field models smooth the interface without explicitly tracking it. Hereby, the solid fraction varies from zero to one over a distance which is typically 4–6 vol elements. In this case a purely Eulerian approach is used to describe the evolution of the solidification field.

Debierre et al. [1] made a first step to extend the phase field approach to model faceted materials for a discrete set of orientations. They focused on capillary effects and neglected the kinetic undercooling of the interface. Their approach consisted in rounding the sharp cusps of the solid-liquid interface free energy plots for equilibrium and non-equilibrium faceted growth. They established a quantitative comparison between phase-field models and sharp interface results

assuming a linear relationship between velocity and undercooling. However the facet's kinetic and furthermore the nonlinear relationship between velocity and undercooling, remain an important step to be included in the phase-field concept.

Taylor and Cahn [2] allowed the phenomenological phase field parameters (such as the gradient energy coefficient and the mobility coefficient for temporal relaxation) to depend on the direction of the gradient of the phase field parameter. In many continuum theories of interface dynamics the interface between the two phases is regarded as being diffuse rather than sharp, with a nonzero thickness over which properties vary smoothly from one set of bulk values to another. Diffuse interface theories permit to use a single equation (or a single system of equations) for the interface and both phases. In this case the discontinuities associated with sharp interface, requiring the solution of separate systems of equations for each phase in a moving boundary problem, can be avoided. Phase field methods add one (or more) order parameters, representing some structural changes and smoothing the transitions in all the other variables. They considered that the interface moves by weighted mean curvature and delivered equations for tracking the edges and corners.

Aside of phase field methods, sharp interface approaches use

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cellular automata technique. It is employed to produce realistic looking crystals, especially in the field of snow crystals growth modelling.

Roosen and Taylor [3] presented a model for crystal growth where the interface between liquid and solid is sharp and explicitly tracked. They assumed that the crystal is a polygon having a limited number of normal growth directions. It is not required that curvature is calculated explicitly, only the measure of the lengths of edges is needed. Since the possible normal directions are known, the comparison between edges with parallel normal is enough to know if two parts of the crystal have intersected or not. Computation time is smaller than in phase field models which require many computational volume elements within the "thickness" of the interface. A related limitation is that the method can be used only for crystalline surface energies. Until now a linear relation between the velocity of the edges and the driving force was considered but also new exponential formulae should be simple to model using the crystalline computational model.

Libbrecht, 2008 [4] simulated diffusion limited growth of faceted crystals using Local Cellular Automata (LCA). This technique was applied to the growth of thin plate-like crystals. The snow crystal is the most studied example of strong anisotropic growth. The discussion was focused on the transition from concave to convex growth as the supersaturation increases. The LCA method was found to be able to simulate the lateral propagation of microsteps on a flat surface, which is a key element in faceted growth.

Barrett et al. [5] simulated 2D and 3D snow crystal growth with a new continuum model using a finite-element approximation for the numerical results. They considered a linear relationship between the tip velocity and the supersaturation. The variation of the condensation coefficient was not sufficient for the faceted growth but produce either plate-like growth or columnar growth. The large effect of the surface energy anisotropy, on the faceted crystal growth was put in evidence. Because many parameters in models for crystal growth are not known, their validation must be done in combination with experimental results.

Libbrecht [6] aimed to develop a basic numerical model to investigate the diffusion-limited growth of ice crystals from water vapour. The model incorporates the cellular automata (CA) technique, presented by Gravner and Griffeath, [7] but uses a more physically realistic treatment of the surface attachment kinetics and of the Gibbs-Thomson effect. The CA numerical model was applied to a 2D cylindrically symmetric crystal growth of relatively simple growth morphologies. Quantitative results were produced, using known diffusion rate, initial sizes and morphologies, for comparison with laboratory crystal measurements. In addition to the morphology the growth rates of the crystal, were nicely reproduced by the model crystal.

Reuther and Rettenmayr [8] presented a review of perspectives of cellular automata for the simulation of dendritic solidification. This method was extensively used for dendritic solidification and less for facetted crystals growth. The problem of the grid anisotropy superposes the strong physical anisotropy encountered in crystals facets and therefore impedes the simulation of this microstructure. The main advantage of the CA method is the lower computational effort.

Most of the crystal growth processes involve fluid flows, such as flows in the melt, solution or vapour. Modelling of faceted growth with flow is mainly found in the field of Czochralski single crystal growth, which is the most important method of growing Silicon crystals for the electronic industry. The melt flow inside the crucible during the Czochralski growth is complicated, thus represents the most difficult part of the modelling. Interactions between buoyancy, surface tension, centrifugal and Lorentz forces were considered in the models of Chen et al. [9]. Initially only a thermo-capillary flow model was considered, later a turbulent flow model was included and last also a magnetohydro-dynamic flow model was applied.

V.V. Kalaev et al. [10] made an attempt to develop an universal numerical model for the optimisation of the Czochralski crystal growth. Global heat transfer (heat transfer in solid parts, and radiative heat exchange between the solid and the melt), inert gas flow and turbulent melt convection, were considered together with a self-consistent computation of the melt-crystal interface, using an unstructured grid. Zaidat team also has developed a global 3D numerical model for the Kyropoulos crystal growth of silicon single crystal [11]. They took into consideration the flow velocity in the furnace, the multiple aspects of the heat transfer (conduction, radiation and convection) and the solidification from Si melt as accounted in the liquid fraction model of Fluent, based on the enthalpy-porosity method. The numerical model was found to be useful to give the operating conditions for a symmetric growth. Their approach (enthalpy-porosity method) assume thermodynamic equilibrium, therefore it cannot model any faceted crystal growth.

Another example of semiconductor material is the GaN crystal, growing in the presence of ammonia water solution (amonothermal growth) under high temperature (200 °C to 500 °C) and pressure (1 to 4 kbar). Chen et al. [12] modelled the 3D flow during the ammonothermal growth using fluid dynamics, thermodynamics and heat transfer and studied the effect of particle size on flow pattern and temperature distribution in the autoclave. A finite volume method was used to solve the governing equations.

Silicon carbide is produced by chemical vapour deposition (CVD) or physical vapour transport (PVT) techniques and represents the third generation of semiconductors used in electronics. In this physical vapour transport growth mechanism, the Stefan flow model based on a flow-kinetics theory was proposed. Chen et al. [13,14] proposed a 1D and a 2D flow kinetics theory for the vapour growth and calculated the concentration distribution and flow field inside the crucible together with the grow rate. Many other growth models were developed, considering electromagnetic field, heating via induction, radiation or conduction heat transfer and growth kinetics. Hofmann et al. [15,16] simulated the flow field in a SiC growth system caused by buoyancy effect, using a finite volume scheme called FASTEST. Pons et al. [17,18] developed a finite element code to predict the electromagnetic field and temperature distribution during the SiC growth.

Demange et al. [19] proposed a modified phase field model able to reproduce complex dynamics of snowflake growth. Two growth regimes were observed. The growth during the first regime was lower than the diffusion. In the second regime the growth follows a linear function. The transition between the two regimes was found to correspond for the horizontal growth to the full faceting of the prismatic face which causes growth to decelerate and for the vertical growth to the formation of a vicinal surface at the needle tip which accelerates the growth. Continuum fluid dynamics could be easily introduced in the model to simulate the air flow around the snowflake.

The growth rate of a triangular shaped crystal was predicted using a hydrodynamic model where the diffusion was taken into account for many flow velocities [20]. Different crystals facets, chosen arbitrarily from 90° to -18° with respect to the flow direction were considered. The main parameter that influences the final crystal shape is the angle between the face and the flow direction. The growth rate decreases downstream along the facets creating an elongated shaped crystal, slightly deviated with respect to the flow direction. The model was applied to a natural calcite crystal and in 2D the results are in good agreement with the observed shapes [21]. The crystal facets with the lowest angle with respect to the flow direction have the highest growth rate and the facets with the largest angle with respect to the flow direction have the highest growth rate.

Three-dimensional phase-field simulations of a silicon crystal in isothermal melt were performed [22]. Both anisotropies of the solidliquid surface energy and of the kinetic attachments were considered, in three different cases, but only when the two effects were considered good agreement was found with experimental results. This study is based on previous experimental, numerical and analytical results [1,23–26].

When the anisotropy of the interface does not play a too important role, both sharp interface tracking and phase field methods give



Fig. 1. Different interface stages during faceted growth. Lateral propagation of a numerical kink.

excellent predictions for dendritic solidification. In opposite, properties of each face can be very different from the neighbouring faces. Thus large jumps in thermodynamics, kinetics and surfaces energy exist at the corners of a crystal, which could lead to strong modelling and computational difficulties, especially for models that smooth the interfaces (phase field).

Based on these issues we have chosen to further develop a sharp interface tracking model to tackle the problem of faceted growth under hydrodynamic condition. The main differences with models described in literature are:

- (1) Interface kinetics, together with solute transport by diffusion and convection are fully taken into account.
- (2) Propagation of kinks is expressively modelled by differentiating normal and lateral growth velocities at each faces.
- (3) Positions of the corners are shifted according to growth of adjacent faces.
- (4) Hydrodynamic is computed with a two-phase model using a penalty method to model the presence of growing obstacles (crystals).

The model is applied to the growth of an isothermal hexagonal crystal in a supersaturated liquid zinc alloy. Modelling of the crystals formation at the macroscopic scale in a liquid zinc bath, was made by Reiss et al. [27].

To illustrate the influence of the flow direction and magnitude, the crystal is chosen to be isotropic, i.e. the interface kinetics is assumed the same for all crystallographic directions. It will be shown that the presence of the flow can lead to di-symmetrisation of the crystal shape, just as difference in interface kinetics between faces can do.

2. Growth model

2.1. Interface kinetics

The theory of solidification proposes different interfacial kinetics depending on different growing mechanisms at the interface. The dendritic structures have a linear interfacial kinetic growth law, where the faceted crystals show a quadratic or an exponential kinetic growth law.

The kinetic growth rate can be expressed using the interface undercooling ΔT_i or the interface supersaturation ΔC_k . The interface supersaturation can be defined as the difference between the liquid concentration at the interface C_l^i and the equilibrium liquid concentration C_l^{eq} :

$$\Delta C_K = C_l^i - C_l^{eq} \tag{1}$$

The different kinetic growth laws can therefore be written as follows:

$$V_n = K_1 \Delta C_k \tag{2}$$

for dendritic growth,

$$V_n = K_2 (\Delta C_k)^2 \tag{3}$$

for spiral growth around an unsaturable defect (screw dislocations, twins),

$$V_n = K_3 \exp\left(-\frac{K_4}{\Delta C_k}\right) \tag{4}$$

for surface nucleation and growth layer by layer.

In the Eqs. (2)–(4), K_1 to K_3 are kinetic coefficients which have different values depending on the crystallographic directions and the type of growth and K_4 is an energy factor. V_n represents the growth velocity perpendicular to the interface (normal velocity).

Besides V_n , for faceted crystal growth, we should also consider a lateral velocity V_{lat} parallel to the surface, at which a kink is filled by atoms. To efficiently build a facet this lateral growth must be much faster than the normal growth. For rough interfaces with large number of kinks (non-faceted growth), the laws governing the lateral growth are represented by the action of a macroscopic curvature undercooling. The numerical resolution of the curvature requires a sufficiently small volume element. However, for faceted growth with limited number of kinks, whose thickness is in the order of atoms, the macroscopic approach using interfacial tension results in an enormous and probably unphysical curvature undercooling.

The curvature undercooling is a macroscopic concept. If we apply it to the curvature of a kink (few Angstroms) we obtain 100 to 1000 K undercooling. The kinetics for a kink is indeed much faster than for a plane interface, its interface concentration (see Eq. (6)) can be considered to be much closer to thermodynamic equilibrium than that of the planar facet. We propose here to mimic the behavior of the kink propagation on the macroscopic mesh. Whenever a numerical "kink" appears on the surface of a facet (Fig. 1), a lateral growth law V_{lat} is applied.

2.2. Solidification growth model

Let us consider one facet of a crystal, the solute balance equation at the interface can be written as follows:



Fig. 2. Schematic representation of a growing crystal as considered in the numerical model, with corner particles (red), facet particles (green). The black arrows represent the resultant growth velocity vectors. (a) initial crystal; (b) constant growth; (c) asymmetric growth: left facet doesn't grow, top facet has the fastest growth and the right facet has a slower growth. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

$$V_{n/lal}C_l^i(1-k_0) = D_s \frac{\partial C_s}{\partial n} \Big|^i - D_l \frac{\partial C_l}{\partial n} \Big|^i$$
(5)

where *n* is the norm of the solidification interface, $V_{n/lat}$ is the normal or lateral growth velocity of the interface, C_l^i and C_s^i are the concentrations of liquid and solid phases at the interface, $k_0 = \frac{C_s^i}{C_l^i}$ is the partition coefficient. On the right hand side the first term represents the diffusion in the solid (with D_s the solute diffusivity in the solid phase, neglected here). The second term represents the diffusion in the liquid (with D_l the solute diffusivity in the liquid phase) which is controlled by hydrodynamics.

The concentration at the facet interface in the liquid C_l^i can be expressed as:

$$C_l^i = C_0 + \frac{1}{m_l} (T^{eq} - T_0 + \Gamma Q f(\varphi, \theta)) + \Delta C_K$$
(6)

where C_0 is the initial liquid concentration, T^{eq} is the equilibrium temperature, T_0 is the initial temperature, m_l is the liquidus slope given by the phase diagram, Γ is the Gibbs-Thompson coefficient, Q is the curvature, $f(\varphi, \theta)$ is a function depending of the orientation and the shape of the crystal. The kinetic supersaturation ΔC_K at the interface is calculated according to the physical growth mechanism (here Eq. (3)) and it is only considered for normal growth (V_n) of facets, not for kinks growth (V_{lat}) , when $\Delta C_K = 0$. In other words with Eq. (5) we will calculate either V_n or V_{lat} . The concentration gradient must be calculated according to the presence of kink: a) for cells with kink (V_{lat}) the concentration gradient is calculated assuming equilibrium at the interface $(\Delta C_K = 0)$; b) for cells without kink (V_n) the concentration gradient is calculated including the kinetic supersaturation $\Delta C_K = \sqrt{\frac{V_n}{K_n}}$.

The lateral velocity results from the cancelation of the interface kinetics from the interfacial concentration. Real kink velocities are much faster than this lateral velocity. However in the present investigation, the lateral velocity was found sufficiently large (compared to normal velocities) to keep every facets almost perfectly flat.

The curvature can be expressed as a function of the solid fraction field with the following equation:

$$Q = \frac{2\partial_x f_s \partial_y f_s \partial_{xy} f_s - (\partial_x f_s)^2 \partial_{yy} f_s - (\partial_y f_s)^2 \partial_{xx} f_s}{[(\partial_x f_s)^2 + (\partial_y f_s)^2]^{3/2}}$$
(7)

The function $f(\phi, \theta)$ can have different forms. For a cubic crystal shape it can be written as follows:

$$f(\phi, \theta) = 1 - 15\varepsilon \cos(4(\phi - \theta)) \tag{8}$$

$$\phi = \arccos\left(-\frac{\partial_x f_s}{[(\partial_x f_s)^2 + (\partial_y f_s)^2]^{1/2}}\right)$$
(9)

2.3. Hydrodynamic and transport of solute

The two-phase volume-averaged Navier-Stokes and mass conservation equations are:

$$\frac{\partial}{\partial t}(\rho_l f_l \vec{u}_l) + \vec{\nabla}(\rho_l f_l \vec{u}_l * \vec{u}_l) = -f_l \vec{\nabla} P + \vec{\nabla}(\mu_l f_l \vec{\nabla} \vec{u}_l) - A \frac{f_s}{f_l + \varepsilon} \vec{u}_l$$

and $\vec{\nabla}(f_l \vec{u}_l) = 0$ (10)

where f_l is the liquid fraction, f_s is the solid fraction, ρ_l is the liquid density, P is the static pressure, μ_l is the liquid viscosity, u_l is the liquid phase velocity and the last term in the first equation represents a penalty factor to force the flow around the growing crystal. The numerical coefficient A should be adjusted according to the strength of the incoming flow. The volume-averaged solute conservation equation used is:

$$\frac{\partial}{\partial t}(f_l\rho_l C_l) + \vec{\nabla}(f_l\rho_l \vec{u}_l C_l) = \vec{\nabla}(f_l\rho_l D_l \vec{\nabla} C_l) - C_s \frac{\partial f_s}{\partial t}$$
(11)

2.4. CA model and front tracking method

The variation of solid fraction (solidification) in a cell of index i, is related to the neighbouring cells j:

$$\frac{\partial f_s^i}{\partial t} = V_n * \lambda_i + \min(V_{lat}, \ \lambda_i / \Delta t) * \sum_j \max(d_j - d_i, 0)$$
(12)

where λ_i is the segment of the interface, Δt the time step, and d_i , d_j the thickness (normal to the interface) of the solidified part of the cell (see Fig. 1).

The solidification model simulates directly the envelope of the crystal with a cellular automaton model. The envelope of the crystal is tracked in a Lagrangian way. With this method, all crystallographic directions of crystal growth can be correctly modelled, for any Eulerian mesh orientation. The envelope of the crystal was separated in corners and facets. Only cells with a solid faction $0 < f_s < 1$ contain envelope tracers. Facets (and associated kinks) grow according to local thermodynamics and kinetics, while corners are simply advected according to adjacent facets. Fig. 2 shows a schematic representation of the growth of an initial crystal (blue hexagon, Fig. 2a) placed at the desired position on the grid. Fig. 2b represents a zoom of the left corner of the

hexagon (see black square in Fig. 2a). In Fig. 2b the preferred growing directions of the crystal are represented by the facets of the hexagon (green tracers). The black arrows represent the resultant growth vectors, which are equal to the sum of normal and the lateral vectors. The corners are tracked via the red tracers. The velocity vector of tracers should advect the interface so that the change of volume fraction includes both normal and lateral growth velocities:

$$V = \frac{\partial f_s}{\partial t} / \lambda_i \tag{13}$$

Two special treatments are applied at cells containing corner tracers. First, advection velocity at corner points is taken as the sum of the advection vectors of the side facets adjacent to it, in second, the source term $\frac{\partial f_s}{\partial t}$ is geometrically calculated according to the new corner position inside the cell.

During the growth a corner moves from one cell to another. To keep the necessary geometrical resolution of the crystal shape, two new facet tracers are created whenever a corner enters a new cell. The advection direction of these facets make a specific angle with the corners direction, this angle is defined by the crystalline orientation (for example $\pi/6$ for a hexagon).

If the advection vectors of the corner tracers (red particles) and of the facet tracers (green particles) are constant, the crystal formed will keep its initial shape and aspect ratio (Fig. 2b). However if the growth vectors on one side are faster than those of the corners, the corners are shifted to that side of the facet (Fig. 2c). Depending on the local conditions, each facet can have its own growth velocity, some facets can be faster than others. If the normal vector velocity in one particle is much faster than in the neighbouring particles then actually the growth will take place laterally and not anymore normal to the facet surface but keeping the facet flat.

3. Results

The facetted crystal simulated in the present paper is the Al_2Fe_5 phase. A microscopic picture of the Al_2Fe_5 phase is given in Fig. 3. It is encountered in the galvanizing industry and these crystals are usually known as top dross particles. Fig. 4 presents the phase diagram at the rich Zn corner, where Fe₂Al₅ phase appears. In Table 1 relevant physical properties and numerical parameters are listed.

For the dross particles no data are available concerning the kinetic growth laws. For sake of simplification a power 2 (quadratic) relation (Eq. (3)), was used for the simulations shown in the present paper, which has the advantage to include only one unknown (the coefficient K_2) compared to the exponential law (Eq. (4)). From the rare available experimental data the K_2 value shown in Table 1, was extracted. For the



Fig. 3. Experimentally observed dross particle (Fe $_2Al_5$ phase). In a 2D cut crystals have a hexagonal shape.



Fig. 4. Phase diagram at the rich Zn corner, at 460 °C, calculated with Mcdermid equation [28] (L stands for liquid phase). The two straight red dotted lines represent the initial Al (0.2 wt%) and Fe (0.02115 wt%) concentrations used in the simulation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1		
Physical properties of li	quid Zink and nu	merical parameters.

Physical properties and numerical parameters	Values
ρ_l	6600 kg/m3
c_p	512 J/kg·K
ĸ	60 W/m·K
μ_l	0.0034 kg/m·s
D_l	1.75·10 ⁻⁹ m ² /s (450 °C)
K_2	$1.14 \cdot 10^{-8} \text{ s}^{-1}$
Γ	$5.5 \cdot 10^{-7}$ (m·K)
Α	10 ⁶
ε	10^{-6}

most studied faceted crystal growth, the Si crystal, recent experiments suggest more a quadratic than a linear kinetic law [29], altough linear kinetics cannot be absolutely excluded, especially for early growth shapes.

The exact value of the Gibbs –Thomson coefficient Γ is unknown, here a relatively large value has been chosen in order to help the interface to remain flat. Naturally the kinetic coefficient K_2 and Gibbs –Thomson coefficient Γ can be different for each facet. This anisotropy will naturally lead at faster growth of some facets (preferred growth direction) and therefore to their disappearance. The dross particle simulated here has an orthorhombic crystallographic orientation. However the microscopic observations (Fig. 3) show a hexagonal shape (in 2D) due to the above mentioned anisotropy, therefore the starting crystal in our simulations has a hexagonal shape. The same values of K_2 and Γ parameters are used for all facets, therefore if the crystal disymmetrizes it will only be the consequence of the kinetics induced by the presence of the flow.

The 2D simulations start with a crystal of 1 μ m size, in a square domain of 300*300 μ m. The crystal is placed exactly in the middle of the domain, aligned with respect to the melt flow direction.

Several situations were simulated: (i) without flow, the growth will be controlled only by diffusion and kinetics; (ii) flow is considered with relative high melt velocities from 7.2 cm/s to 1.44 m/s. First situation reproduces the conditions of faceted crystal growth on a stationary support and the second situation approaches the conditions of a faceted crystal growth on a moving support.



0.011 wt% Fe

0.02 wt% Fe

Fig. 5. Crystal (dark blue) evolution without flow and the corresponding Fe concentration field in the liquid around the crystal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 6. Crystal (dark blue) evolution under 0.072 m/s melt flow and the corresponding Fe concentration field in the liquid around the crystal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.1. Without melt flow

Fig. 5 presents the simulation results of the crystal growth without

melt flow. It can be observed that the diffusion layer is thick and uniform around the crystal. The tracks of the corner's positions are straight lines. The crystal keeps its symmetric shape during growth, as all facets

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Fig. 7. Crystal (dark blue) evolution under 0.36 m/s melt flow and the corresponding Fe concentration field in the liquid around the crystal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 8. Crystal (dark blue) evolution under 0.72 m/s melt flow and the corresponding Fe concentration field in the liquid around the crystal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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Fig. 9. Crystal (dark blue) evolution under 1.44 m/s melt flow and the corresponding Fe concentration field in the liquid around the crystal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



288 s

Fig. 10. Melt flow around a growing crystal at 288 s. Velocity magnitude (colours) and vectors field (arrows).

have the same kinetics (Gibbs-Thomson coefficient and interfacial kinetic coefficient). This case can be used as reference for the following cases involving several melt flow intensity.

3.2. With melt flow

The flow magnitude imposed at the left boundary limit was varied between 0.072 and 1.44 m/s. The crystal's facets at the left hand side of the pictures, where the melt flow meets directly the crystal are called front facets, by opposition of the facets at the right hand side of the pictures, called back facets. Additionally we define the top and the bottom facets. Figs. 6–9 present the results of the crystal growth at different melt flow velocities.

For all cases presented in Figs. 6–9 the diffusion boundary layer is not similar for the different crystal's facets. At the front facets the boundary layer is much thinner than at the other facets. Moreover with the increase of the melt flow velocity it can be observed that the solute diffusion boundary layer becomes thinner.

Another important fact is that the traces of the corner's positions are not straight lines as in the case of crystal growth without flow. Already at around 80 s (second pictures in Figs. 6 to 9) it can be observed that the crystal starts losing its symmetry; the initial centre point is as "moved to the right". The upwind part of the crystal grows faster as its downwind part.

For the last two cases at 0.72 m/s and 1.44 m/s melt flow, starting with approximately 60 s and respectively 22 s, a back flow was observed, resulting from the occurrence of the so called von Kármán vortices, (Fig. 8b) and Fig. 9b)). For a flow around a hexagon, the von Kármán vortices appear at a critical Reynolds (*Re*) number between 54 and 55 [30]. The dimensionless *Re* number is calculated as follows:

$$Re = \frac{\rho_l U L_2}{\mu_l} \tag{14}$$



Fig. 11. Evolution of crystal growth for different flow velocities. (a) Evolution of crystal width (L_1) ; (b) Evolution of crystal height (L_2) ; (c) Evolution of crystal growth rates in the direction parallel to the melt flow; (d) Evolution of crystal growth rates in the direction perpendicular to the melt flow; (e) Evolution of crystal growth rates in the direction parallel to the melt flow with the width of the crystal (L_1) .

where L_2 is the length of the crystal perpendicular to the flow direction and U is the melt flow velocity.

The corresponding critical *Re* number value, for the above presented simulations, turned out to be approximately 53.

Fig. 10 presents the velocity magnitude of the melt flow around the growing crystal and the corresponding velocity field. The melt flow is forced to go around the crystal and its intensity becomes stronger once reaching the top and bottom facets. Vortices can be observed at the top and bottom facet as well as behind the crystal, where the von Kármán vortices form.

In Fig. 11a) and b) the evolution of the crystal size in time (L_1 is the measured size of the crystal in the direction parallel with the melt flow (crystal width) and L_2 is the size of the crystal in the direction perpendicular to the melt flow (crystal height)) is compared for the different melt flow velocities.

The width of the crystal (L_1) for all cases with melt flow increases faster than the height (L_2) of the crystal. This situation is the result of two facts: (i) the diffusion boundary layer is smaller for the front facets than for the top and bottom ones, therefore the growth at these facets (front facets) is faster; (ii) the back flow due to the von Kármán vortices contributes also to an increase in the growth rate at the back facets.

Furthermore the shape of the crystal is not anymore symmetric, the back facets of the crystal (right hand side of the picture) are growing slower than the front facets (left hand side of the picture). This can be correlated to the boundary diffusion layer, which is much thinner for the front facets than for the back one, therefore the mass transfer is faster at the front that at the back of the crystal.

From Fig. 11a) and b) it can be observed that the crystal size increases almost linearly but at around 75 s a change in slope point can be noticed, which means that the crystal's size increases slower after this point. In Fig. 11c), d) and e) the corresponding growth rates are reported. A general trend can be observed for all cases: the growth rate decreases in time (Fig. 11c), d)) and respectively with the increase of the crystal size (Fig. 11e)). This can be explained by the growth of a cylindrical crystal in 2D, which is scaled with the inverse of its size: $v \propto \frac{1}{L}(C_0 - C_i)$, the growth rate decreases with the increase of the crystal's size [31].

Additionally comparing Fig. 11c) and d) it can be observed that larger is the melt flow velocity smaller is the growth rate decrease. However the decrease of the growth rate in the direction parallel to the melt flow is slightly smaller than the decrease on the growth rate in the direction perpendicular to the melt flow for the same melt flow velocity (Fig. 11c) and d)). The difference between the growth rate in the two direction (parallel and perpendicular to the melt flow) increases with the increase of the melt flow velocity (Fig. 11c) and d)).

From Fig. 11c) and d), it can be noticed that for the case of no melt flow respectively the case of 0.072 m/s melt flow velocity, the crystal grows with almost similar growth rates in both directions (parallel and perpendicular to the flow). The case with 0.36 m/s melt flow starts to present a significant difference between the growth rates in the two directions parallel and perpendicular to the flow (Fig. 11c) and d)). The crystal grows faster in the direction parallel to the flow compared to the direction perpendicular to the flow. For the cases of 0.72 and 1.44 m/s melt flow velocity the difference between the growth in the two directions (parallel and perpendicular to the flow) becomes even larger (Fig. 11c) and d)). This can be explained by the influence of the flow, which will decrease the boundary diffusion layer at the front facets interface and therefore will increase the growth rate. For the back facets the von Kármán vortices will transport fresh Zn liquid with higher Fe concentration from bulk melt and therefore the growth rate is increased.

Furthermore a correlation between the melt flow velocity and the growth rate can be established, larger is the melt flow velocity, faster is the growth rate of the crystal. In all the cases the model was able to keep the facets flat as observed in experiments (Fig. 3).

4. Conclusions

The cellular automaton model employed here was able to simulate the growth of faceted crystals. One single crystal was simulated using a quadratic kinetic growth law, in two scenarios, with and without melt flow. The influence of the flow on the growing crystal and reciprocally the influence of the growing crystal on the melt flow were put in evidence.

The single crystal without melt flow grows slower with a perfectly symmetric shape.

Different melt flow velocities were simulated. Crystal was found to grow faster with the increase of melt flow velocity. The diffusion boundary layer is reduced in presence of melt flow, therefore the mass transfer and consequently growth rate is faster in presence of flow.

The flow is forced to contour the growing crystal. At highest melt flow velocities, the von Kármán vortices appear at the rear of the crystal.

Furthermore, the growth rate of the faceted crystal was found to decrease with its size until a plateau is reached.

The current simulations can only offer a qualitative comparison with the experimental results. More experimental data should be provided in order to estimate the kinetic interfacial coefficient. Effect of the orientation of the flow with respect of the crystal should be studied. Afterwards the current model could be applied to simulate the competition of many crystals growing over a solid surface (so called "buildup" growth).

CRediT authorship contribution statement

Mihaela Stefan-Kharicha: Writing - original draft, Visualization, Investigation. Abdellah Kharicha: Conceptualization, Supervision, Writing - review & editing. Kader Zaidat: Conceptualization. Georg Reiss: Writing - review & editing. Werner Eßl: Writing - review & editing. Frank Goodwin: Writing - review & editing. Menghuai Wu: Conceptualization. Andreas Ludwig: Conceptualization. Claudia Mugrauer: Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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