International Journal of Thermal Sciences 126 (2018) 252–262

Contents lists available at ScienceDirect

International Journal of Thermal Sciences

journal homepage: www.elsevier.com/locate/ijts

Mechanical stirring influence on solute segregation during plane front directional solidification

M. Chatelain[a,b,*], V. Botton[c,d], M. Albaric[a,b], D. Pelletier[a,b], B. Cariteau[e], D. Abdo[e], M. Borrelli[e]

[a] Univ. Grenoble Alpes, INES, F-73375 Le Bourget du Lac, France
[b] CEA, LITEN, Department of Solar Technologies, F-73375 Le Bourget du Lac, France
[c] Laboratoire de Mécanique des Fluides et d'Acoustique, CNRS/Université de Lyon, Ecole Centrale de Lyon/Université Lyon 1/INSA Lyon, ECL, 36 Avenue Guy de Collongue, 69134 Ecully Cedex, France
[d] INSA Euromed, Université Euro-Méditerranéenne de Fès, route de Meknes BP 51, 30000 Fez, Morocco
[e] DEN/DANS/DM2S/STMF/LIEFT, CEA Saclay, 91191, Gif-sur-Yvette Cedex, France

ARTICLE INFO

Keywords:
Directional solidification
Segregation
Mechanical stirring
Convection
Silicon purification

ABSTRACT

The present paper focuses on directional solidification processes for photovoltaic silicon purification. The use of a mechanical stirrer in the melt to enhance impurity segregation is investigated through numerical simulations. The 3D forced convection flow is resolved in a transient regime thanks to a sliding mesh approach. The hydrodynamic model is coupled to a solute transport simulation in a quasi-steady approximation (i.e. with constant liquid height). Velocity measurements are performed by Particle Image Velocimetry on a water model in order to validate hydrodynamic simulations. Numerical results show that an efficient segregation can be achieved, even for high solidification rates, thanks to mechanical stirring. The numerical model provides meaningful insights for process optimization as it correlates the impurity repartition on the solidification front to the stirring parameters. Finally, the numerical segregation results are compared to an analytical model of the solute boundary layer. It is found that the analytical model provides a good estimate of the mean segregation regime from an hydrodynamic simulation of the forced convection flow, which makes it a useful tool for process design.

1. Introduction

1.1. Context

The silicon purification is an important issue for the photovoltaic industry since impurity concentrations have a strong influence on solar cells efficiency [1,2]. In order to reduce costs and environmental impact, specific purification processes, based on metallurgical operations, are developed for photovoltaic silicon [3]. The segregation phenomenon, which occurs during a directional solidification process, is an efficient way to remove metallic impurities which are usually far less soluble in solid silicon than in liquid silicon and thus feature low partition coefficients [4]. An intense convection in the melt is, however, required to ensure efficient segregation, especially if high growth rates are targeted, for productivity sake. Various stirring techniques can then be used during directional solidification processes to control fluid flow and improve material quality.

Electromagnetic stirring is already widely used for Czochralski processes, in order to tailor convection and control dopants and oxygen incorporation [5–7]. This technique is also investigated for directional solidification processes [8–11]. Nevertheless, skin effect is expected to limit the velocity magnitude in the bulk of the melt for large ingots [12]. Mechanical stirring is an interesting solution allowing high velocities and strong turbulence in the melt, which is desirable for an efficient segregation in large industrial ingots (crucible width in the order of 1 m). This technique is widely used in chemical industries for mixing operations [13,14] and for some metallurgical processes like aluminum degassing [15–17]. This solution was recently investigated for silicon directional solidification [12,18]. Other stirring techniques can also be cited, such as acoustic stirring [19–21] or the use of steady magnetic fields combined with electrical current injection [22]. The present paper focuses on a pilot scale directional solidification process (crucible width 38 cm) using an axial impeller to ensure an efficient segregation of metallic impurities.

1.2. State of the art

The simulation of fluid convection flow and solute transport in the melt represents an efficient optimization tool for the silicon purification process by segregation. In the literature, the simulation of rotational

---

[*] Corresponding author. Univ. Grenoble Alpes, INES, F-73375 Le Bourget du Lac, France, CEA, LITEN, Department of Solar Technologies, F-73375 Le Bourget du Lac, France.
E-mail address: m.chatelain.m@gmail.com (M. Chatelain).

https://doi.org/10.1016/j.ijthermalsci.2017.12.024
Received 30 August 2017; Received in revised form 27 November 2017; Accepted 21 December 2017

Nomenclature

Δ	convecto-diffusive parameter (−)
δ	solute boundary layer thickness (m)
μ	dynamic viscosity (Pa.s)
μ_t	turbulent dynamic viscosity (Pa.s)
ν	kinematic viscosity (m^2 /s)
ν_t	turbulent kinematic viscosity (m^2/s)
ρ	density (kg/m^3)
τ	wall shear-stress (Pa)
$\vec{u}$	fluid velocity (m/s)
B	dimensionless segregation parameter (−)
c	distance from impeller to solidification front (m)
C_L	solute mass fraction in the liquid (−)
C_S	solute mass fraction in the solid (−)
D	molecular diffusivity (m^2/s)
d	impeller diameter (m)
D_t	turbulent diffusivity (m^2/s)
Fq	dimensionless pulsation (−)
H	liquid silicon height (m)
k_0	segregation coefficient (−)
k_{eff}	effective segregation coefficient (−)
L	crucible width (m)
N	rotational speed (rpm)
p	pressure (Pa)
Re_d	Reynolds number (−)
Sc	Schmidt number (−)
Sc_t	turbulent Schmidt number (−)
V_I	interface velocity (m/s)

impellers has been widely investigated, especially for chemical processes applications. Different methods have been proposed in order to predict fluid motion and turbulence properties in stirred tanks. Readers are referred to Aubin et al. [13] and Brucato et al. [14] for a review of the different methods. At first, empirical models, like Impeller Boundary Conditions (IBC) or momentum source terms models, were proposed. Such simplified approaches rely on experimental data and provide no information on the flow in the impeller region. Then, with the increase of computational capacities, more sophisticated methods, describing the exact geometry of the impeller, were proposed. The Multiple Frames of Reference (MFR) approach was first introduced by Luo et al. [23]. In this approach, the computational domain is divided in two zones by a cylindrical interface encompassing the impeller. In the impeller region the flow is computed in a frame rotating with the impeller, and in the outer region it is computed in a fixed reference frame. Aubin et al. [13] explain that this method is a steady-state approximation which is valid when the distance between the tank walls and the impeller blades is sufficiently large to assume that the tank geometry has no influence on the flow in the impeller region. For a complete transient resolution of the flow, a sliding mesh technique must be used to simulate the impeller rotation. This approach requires more computational means, but involves no assumption on the impeller flow and its interaction with the tank. In these problems, turbulence description often represents a major issue. As a matter of fact, turbulence has a direct influence on mixing processes. Recent studies focus on this aspect and investigate the potential of advanced turbulence models, like Large Eddy Simulation (LES), to enhance stirring models performances [24,25]. Once again, these developments are made possible by the increase of computational capacities.

Experimental characterization of stirred tanks is also widely investigated in the literature [26]. The aim is to provide physical understanding of the transfer processes and to improve and validate numerical models. Regarding velocity field measurements, optical techniques are usually preferred. For instance, Laser Doppler Velocimetry (LDV) is often used to produce precise measurements near the impeller because it provides a high spatial and temporal resolution [26]. Alternatively, Particle Image Velocimetry (PIV) is an efficient technique which gives access to the velocity field on a complete plane of the flow. This aspect makes it a very suitable technique for the characterization of global flow structure in stirred tanks [27–29]. Metallurgical processes can also be investigated thanks to experimental models, providing that right similarity conditions are defined. As an example, Camacho-Martínez et al. [15] investigated a pilot system of aluminum degassing using a full scale water physical model. PIV measurements were performed to determine flow patterns and turbulence characteristics in the stirred tank.

On the other hand, several numerical studies are dedicated to the impact of convection flow on directional solidification and solute segregation [8,9,30–32], but few involve a mechanical stirrer. Dumitrica et al. [12] performed a numerical investigation of mechanical stirring influence on a silicon directional solidification process. Authors used momentum source terms to reproduce the action of a cylindrical stirrer (tangential velocity component). The aim of the study was to highlight the influence of the rotation speed on the solid/liquid interface deflection. Analytical approaches are also developed in order to estimate the solute boundary layer from the knowledge of the melt convection flow. Recently, analytical models of the solute boundary layer have been proposed by Altenberend et al. [33] in the case of 1D unbounded turbulent parallel flows and Garandet et al. [34] for a 2D laterally confined flow. This last model is derived from a scaling analysis procedure previously introduced for the study of segregation under natural convection in Czochralski and horizontal Bridgman growth configurations [35]. The aim of this analytical model is to provide an estimate of the segregation regime, without performing a complete simulation of the segregation problem. The finality is to avoid a numerical resolution of the solute boundary layer, which can be demanding in computational means. An extension of this model for fully turbulent configurations, with significant effects of the turbulent transport inside the solute boundary layer, has been proposed in a recent publication [36]. Up to now, this model has been tested in a canonical reference case, namely the 2D lid driven cavity, both in quasi-steady and transient regimes. Kaddeche et al. [37] also compared the analytical model to numerical and experimental results of segregation under natural convection in horizontal Bridgman configurations.

1.3. Objective of the study

The aim of this work is to propose numerical and analytical tools for the study of solute segregation under mechanical stirring. The present study focuses on the application of an axial impeller for a silicon purification process. In this frame of work, fluid flow must be precisely described in order to get a realistic picture of the solute transport in the liquid phase. 3D transient simulations of the flow generated by the axial impeller are, therefore, performed using a sliding mesh approach. An experimental characterization of the velocity field is performed by PIV measurements on a water experiment model for the validation of the stirring simulations. The hydrodynamic simulations are coupled to a solute transport computation. Segregation is an inherently transient phenomenon since solute rejection into the liquid phase is generated by the displacement of the solidification front. The representative time scale of the solidification process is, however, much larger than the time scale associated with the rotation of the impeller. A quasi-steady assumption is thus adopted regarding the solidification process, as formerly proposed in several studies [34,38–42]. This numerical model is used to investigate the influence of stirring parameters on the solute boundary layer at the growth front. Finally, numerical segregation