Journal of Crystal Growth 409 (2015) 89–94

Contents lists available at ScienceDirect

Journal of Crystal Growth

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

On the effect of natural convection on solute segregation in the horizontal Bridgman configuration: Convergence of a theoretical model with numerical and experimental data

S. Kaddeche [a,*], J.P. Garandet [b], D. Henry [c], H. Ben Hadid [c], A. Mojtabi [d]

[a] University of Carthage, Institut National des Sciences Appliquées et de Technologie (INSAT), Laboratoire de Recherche Matériaux, Mesures et Applications, B.P. 676, 1080 Tunis Cedex, Tunisia
[b] CEA, LITEN, Department of Solar Technologies, National Institute of Solar Energy, F-73375 Le Bourget du Lac, France
[c] Laboratoire de Mécanique des Fluides et d'Acoustique, CNRS/Université de Lyon, École Centrale de Lyon/Université Lyon 1/INSA de Lyon, ECL, 36 Avenue Guy de Collongue, 69134 Ecully Cedex, France
[d] Institut de Mécanique des Fluides de Toulouse, UMR CNRS/INP/UPS 5502, UFR MIG, Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex, France

ARTICLE INFO

Article history:
Received 1 September 2014
Received in revised form
2 October 2014
Accepted 4 October 2014
Communicated by: Chung wen Lan
Available online 12 October 2014

Keywords:
A1. Convection
A1. Directional solidification
A1. Segregation
A2. Growth from melt

ABSTRACT

The effect of natural convection on solute segregation in the horizontal Bridgman configuration is studied. The objective is to check whether a single non-dimensional number, based on the fluid flow induced interface shear stress, is able to capture the physics of the mass transport phenomena. A number of heat and mass transfer numerical simulations are carried out in the laminar convection regime, and the segregation results are found to be in good agreement with the predictions of the scaling analysis. At the higher convective levels relevant for the comparison with existing experimental data, a direct computation of the segregation phenomena is not possible, but numerical simulations accounting for turbulence modeling can provide the interface shear stress. With this procedure, a good agreement between the experimentally measured segregation and the predictions of the scaling analysis is again observed, thus validating the choice of the interface shear stress as a key parameter for the segregation studies.

1. Introduction

In melt growth technologies, solute or impurity segregation often represents an important issue, e.g. for the control of solidification in concentrated semiconductor alloys [1] or for the purification of upgraded metallurgical grade Si feedstock in photovoltaic applications [2]. For such an issue, the role of both Fickian diffusion and convection has been widely recognized in the past, but a global understanding is still missing. As a matter of fact, the global heat, momentum and mass transport problem features a variety of length scales, particularly due to the existence of thin solute boundary layers in the vicinity of the solidification interface which often prevents an accurate global numerical modeling of the growth configuration. Therefore models allowing to somehow decouple species transport from the heat and momentum transport problems can be very useful. In such a perspective, order of magnitude analyses can provide interesting insights, particularly if the objective is primarily to determine whether impurity transport is mainly driven by convection or by diffusion. As a matter of fact, it must be understood that such approaches cannot be expected to be quantitatively accurate, but they can provide scaling laws and as such useful insights in the physics of the transport phenomena.

Attempts in this direction are not new, starting from the pioneering work of Burton et al. [3], later on referred to as BPS, in their model Czochralski configuration. In this pioneering work, BPS managed to relate the characteristics of the forced convection flow to the effective partition coefficient thanks to a newly introduced convecto-diffusive parameter. This pioneering work was later on refined by Wilson [4], who proposed a scientifically sound definition for the solute boundary layer thickness and the convecto-diffusive parameter. In addition to Czochralski growth, this approach proved very useful for the interpretation of the numerical simulation results of Kaddeche et al. [5] in the horizontal Bridgman configuration.

On a related line of thought, Ostrogorsky and Müller [6] proposed a model based on a mass balance and the related solute fluxes across the growth interface to yield the effective partition coefficient and the boundary layer thickness. In a couple of recent papers, Ostrogorsky [7] relied on correlations for the convective

* Corresponding author.
E-mail addresses: Slim.Kaddeche@insat.rnu.tn, slimkaddeche@yahoo.fr (S. Kaddeche).

http://dx.doi.org/10.1016/j.jcrysgro.2014.10.009
0022-0248/

mass transport coefficient in various fluid flow configurations to derive estimates of the partition coefficient. A common feature of all the above literature is that knowledge of some external features of the involved fluid flow is necessary as an input in the mass transport problem. As a consequence the results are presented as a function of various non-dimensional groups that a priori characterize the convection problem.

Such is not the case in the recent work by Garandet et al. [8] where the authors proceed to define the local velocity field based on the interface shear stress induced by the motion of the fluid. As such, the physical nature of the convective driving forces does not explicitly appear in the theoretical frame, which can, as a consequence, be considered universal in nature. It should of course be stated that the interface shear stress may not be fully familiar to the experimenter, but in the frame of an approach where numerical simulations are carried out for heat transfer and fluid flow, it is readily available as a result of the computations.

In any case, comparisons with numerical results obtained in the lid driven cavity configuration support the validity of the theory [8] and the ability of the scaling analysis to capture the physics of the segregation phenomena. However, in part due to the fact that lid driven convection is rarely encountered in crystal growth devices, the necessity of further tests for this model had been mentioned [8]. In this respect, the horizontal Bridgman configuration presents a number of advantages, due to a well-defined convective driving source, and more important to the existence of a relatively large and reliable numerical [5] and experimental [9–10] data base.

In Section 2, we will first briefly outline the theoretical model along with the procedures involved in the determination of the numerical and experimental data base that will be used for the comparisons. We will then proceed in Section 3 to the presentation of the results, along with a discussion of the validity of the model.

2. Background and procedures

2.1. Model formulation

Our purpose here is only to briefly recall the outline of the procedure. For more details the interested reader is referred to Ref. [8]. Our starting point is the convecto-diffusive mass balance equation, which governs the concentration C of an impurity or a dopant (expressed here as mass fraction) in a frame moving with the solid–liquid interface at a rate V_I along the Z-direction

$$\partial C/\partial t+(\mathbf{V}.\nabla)\, C = D\nabla^2 C+V_I\partial C/\partial Z, \tag{1}$$

$\mathbf{V}$ and D respectively standing for the convective velocity, solution of the Navier–Stokes equations, and the impurity or dopant diffusion coefficient. Closed form analytical solutions to Eq. (1) exist only in rare cases, such as diffusion controlled growth ($\mathbf{V}=0$), thus requiring the recourse to numerical simulations or simple order of magnitude analyses, as carried out in [8]. The model is based on approximate expressions in a two-dimensional representation for the components of the convective flow parallel and normal to the interface, denoted respectively as U (along the vertical coordinate X) and W (along the horizontal coordinate Z). More specifically, it is supposed that away from the cavity lateral walls, U and W can be written as follows:

$$U(Z)\sim(\tau/\eta)\, Z, \quad W(Z)\sim(\tau/\eta H)\, Z^2, \tag{2}$$

where τ represents the interface shear stress, generally defined as $\tau=\eta\left(\partial v_t/\partial x_n\right)_I$ where v_t is the tangential velocity, x_n is the normal direction and the subscript I indicates an evaluation at the interface (here, with our notations, $\tau=\eta\left(\partial U/\partial Z\right)_I$), η is the dynamic viscosity of the fluid, H is a characteristic macroscopic dimension of the solid–liquid front and Z is the distance from the point of interest to the interface. At this point, it should be mentioned that the concept of 'interface shear stress' may appear questionable since, from a physical standpoint, the key physical parameter defining the flow field is rather the gradient of the tangential velocity in the direction normal to the interface. Nevertheless, in Newtonian fluids (as those considered in the present work), this quantity is linearly related to the interface shear stress and can be written as τ/η as expressed in Eq. (2).

In addition, the concept of interface shear stress is commonly used in the turbulence literature in the context of wall bounded shear flows, resulting in the fact that as mentioned earlier, the values of the interface shear stress are readily available as a result of the numerical simulations in turbulent flow conditions in standard commercial codes. Finally, from an experimental standpoint, it should also be stated that a number of techniques have been developed for the measurement of wall shear stresses [11]. In view of all these arguments, reference will be made to interface shear stress all through the paper, even though it should be remembered that a presentation of the results in terms of normal velocity gradients would also be possible.

In any case, as discussed in [8], it is expected that the expressions given by Eq. (2) will be adequate in both laminar and turbulent convective configurations if in the latter case, U and W are meant to represent the components of the Reynolds averaged velocity field. The scaling analysis then allows deriving the value of the convecto-diffusive parameter Δ (namely the dimensional solute boundary layer thickness δ normalized by D/V_I) as a function of a 'universal' nondimensional group given as

$$B=\tau D^2/{V_I}^3\eta H. \tag{3}$$

The analytic expression obtained is given in Ref. [8]. For the sake of completeness, it should be recalled that the convecto-diffusive parameter Δ is of paramount importance in segregation problems, since it can be univocally related to the thermodynamic and effective partition coefficients k and k_{eff} according to the formula $k_{eff}=k/(1-(1-k)\Delta)$ [4].

2.2. Numerical procedures

Our objective in this section is again only to outline the numerical procedures used in the present work. We actually relied on two distinct codes, a two dimensional in-house program for a detailed comparison with the predictions of the scaling analysis in laminar fluid flow configurations, and the commercial software Fluent, which was used for the derivation of the interface shear stress in turbulent conditions in order to test the scaling analysis against the experimental data.

Regarding the in-house code [5], the governing equations were solved in a vorticity–stream function formulation using an alternating direction implicit (ADI) technique, with a finite-difference method involving forward differences for time derivatives and Hermitian relationships for spatial derivatives, resulting in a truncation error in $O(\Delta t^2, \Delta X^4, \Delta Z^4)$, i.e. of second and fourth orders in time and space, respectively (see Hirsh [12] and Roux et al. [13]). The mesh used to solve the problem was generated by a technique initially proposed by Thompson [14]. The node density is of course larger near the side walls of the cavity, especially in the vicinity of the growth interface. As shown in [5], a 25×101 grid guarantees a sufficient accuracy for such studies. Regarding physical assumptions, only the thermal convection in the Boussinesq approximation was considered, which amounts to assuming that the alloy is sufficiently dilute for solutal buoyancy to be negligible.

A schematic of the problem is shown in Fig. 1. In dimensional form, the parameters of the problem are the cavity width H, length L,