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PARAMETRIC ANALYSIS OF THE RADIAL SEGREGATION IN DIRECTIONAL SOLIDIFICATION OF ALLOYS

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Abstract
In this work, a parametric analysis of the radial solute distribution in crystals grown by the vertical Bridgman method is performed in order to find optimal conditions for growing crystals with improved chemical homogeneity. The analytical formulas from the literature are used to estimate the convecto-diffusive parameter and the radial segregation. The parametric analysis performed by varying the growth rate and the diffusion coefficient can predict the conditions for obtaining a convective regime in the melt. As theoretically shown, the crystals grown in these conditions are characterized by small radial variations of the solute concentration.

1. Introduction

The vertical Bridgman method is widely used in crystal growth of diluted and concentrated alloys. In this method, a melted charge is solidified by pulling the crucible at constant rate in the thermal field produced by a furnace. The quality of crystals grown by directional solidification of doped melts is generally affected by the chemical segregations. As for example, the crystals grown by the vertical Bridgman method are characterized by significant axial and radial segregations. The solute distribution in crystals depends on the diffusive and convective transport conditions in melts during the growth process. As shown in a previous numerical work [1] the chemical segregations are significant reduced for a convective regime characterized by a good mixing of the solute in the melt.

In this work, a parametric analysis of the radial segregation is performed in order to find optimal conditions for growing crystals with improved chemical homogeneity.

2. Method

The numerical analysis performed in [1] have shown that chemical homogeneity of crystals grown by vertical Bridgman method can be improved when the solidification occur in conditions of a convective regime in the melt. The convecto-diffusive transport conditions in the melt are described by the convecto-diffusive parameter:

$$\Delta = \frac{V}{D}\delta \tag{1}$$

where δ is the width of the boundary solutal layer which appears near the solid-liquid interface and is characterized by a significant variation of the solute concentration.

A purely diffusive regime is characterized by a parameter $\Delta = 1$ and a convective regime occurs at small values of Δ ($\Delta \le 0.2$). When the solidification occurs under full convective conditions, the solute is well mixed and the crystals are characterized by small variations of the solute concentration on the axial and radial directions. Therefore it is important to find the growth parameters that characterize a convective regime in the melt.

The convecto-diffusive parameter can be estimated by using Kaddeche's formula [2]:

$$\Delta = 13.2 \cdot Pe \cdot (Gr \cdot Sc)^{-1/3} \tag{2}$$

In the above equation, the non dimensional numbers of Peclet (Pe), Grashof (Gr) and Schmidt (Sc) are given by the following expressions:

$$Pe = \frac{V \cdot R}{D}, \quad Gr = \frac{B_T \cdot \delta T \cdot g \cdot R^3}{\nu^2}, \quad Sc = \frac{\nu}{D} \tag{3}$$

where V, R, D, ν, g and β_T are respectively the growth rate, the sample radius, the diffusion coefficient, the viscosity, the gravitational acceleration and the thermal expansion coefficient. The radial variation of the temperature at the interface depends on the interface deflection f and the thermal gradient G_T:

$$\delta T = f \cdot G_T \tag{4}$$

The radial variation of the solute concentration is given by the radial segregation:

$$\delta C = \frac{C_{max} - C_{min}}{C_{av}} \tag{5}$$

where C_{max}, C_{min} and C_{av} are the maximum, minimum and the average concentration in liquid at the interface.

The radial segregation for samples grown under full convective condition is given by the following formula [2]:

$$\delta C = 1.1 \cdot (1-K) \cdot Pe \cdot Gr^{-1/9} \cdot Sc^{-2/9} \qquad (6)$$

In the following, the convecto-diffusive parameter Δ and the radial segregation δC are analyzed as function of the convective intensity given by the Grashof number. [3]

3. Results and Discussions

In the Fig.1a, the parameter Δ is represented as function of the Grashof number for a given growth rate $V = 1\mu m/s$ ($Pe = 0.5$). It can be observed that a convective regime characterized by $\Delta \le 0.2$ is obtained for values of the Grashof number $Gr > 1000$. This suggests a moderate intensity of the convection. As for example, for a GaInSb sample with the interface deflection $f = 0.5mm$ solidified in an external thermal gradient $G_T = 50K/cm$, the Gr number is $Gr = 5000$. Therefore, it is expected to have a convective regime for the growth of these crystals at $V = 1\mu m/s$.

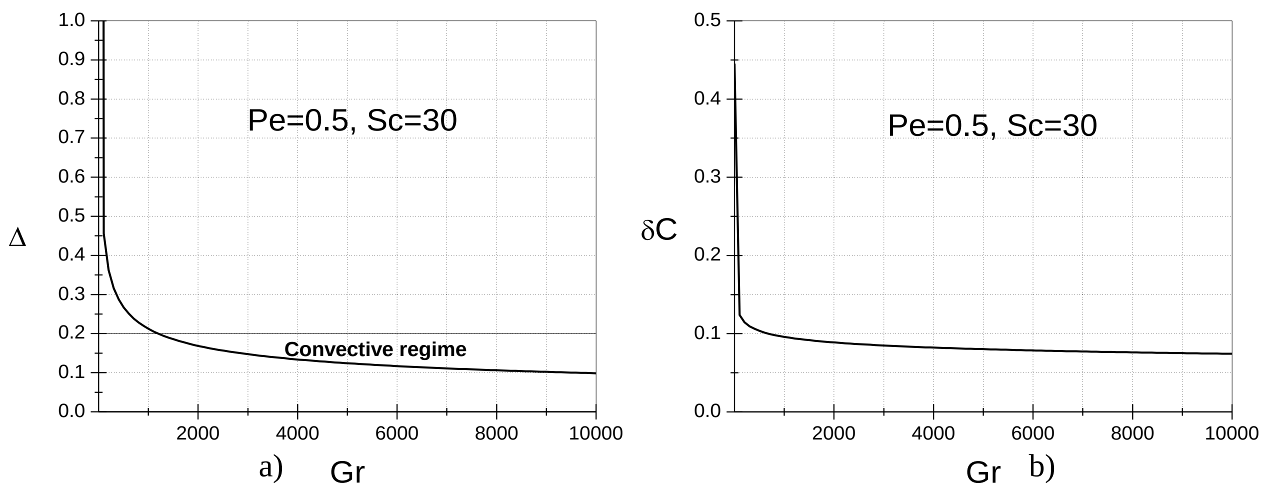


Fig.1 a) Convecto-diffusive parameter Δ as function of the Gr number; b) Radial segregation δC as function of the Gr number. Growth rate $V = 1\mu m/s$

The radial segregation as function of the Gr number is represented in the Fig.1b. It can be observed that the radial segregation has huge values for $Gr < 1000$, but decreases to $\delta C \approx 0.07$ for $Gr > 1000$.

So, the crystals grown at $V = 1\mu m/s$ have small values of the radial segregation because of the convective growth conditions at $Gr > 1000$.

In the Fig.2a the parameter Δ is represented as function of the Gr number at an increased growth rate $V = 2\mu m/s$. The convective regime is obtained at higher values of the Grashof number $Gr > 10000$, which indicate a high level of the convection in the melt. In this case the thermal field can be affected by the flow and the solid-liquid interface would be distorted. That has a negative effect on the crystals because the thermal stresses increase at the interface. As shown in the Fig.2b, the radial segregation increases to values of $\delta C \approx 0.15$ for $Gr > 10000$.

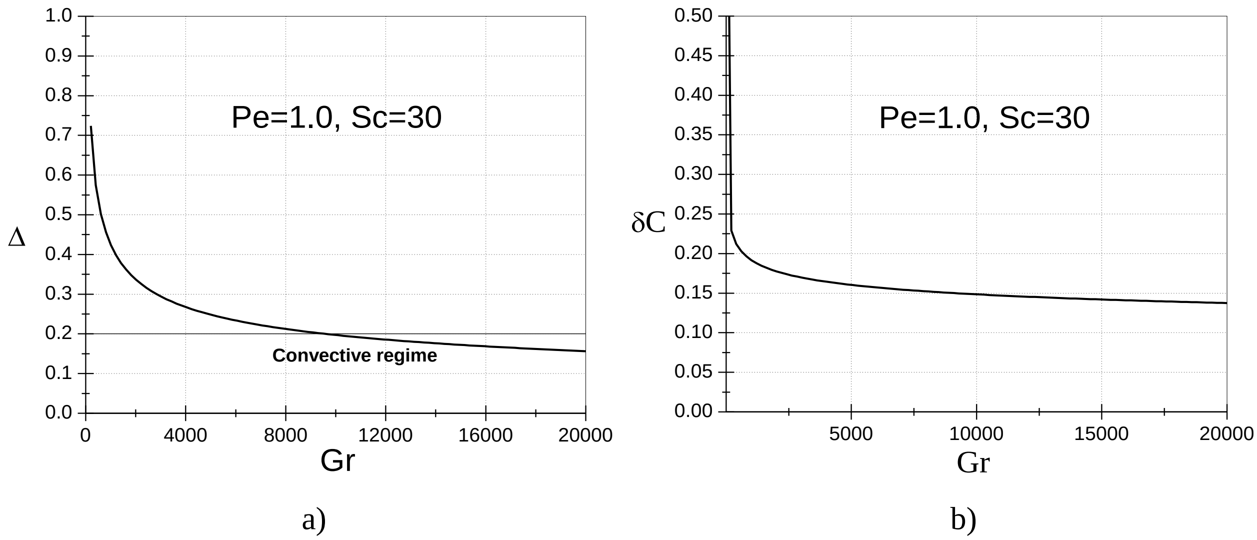
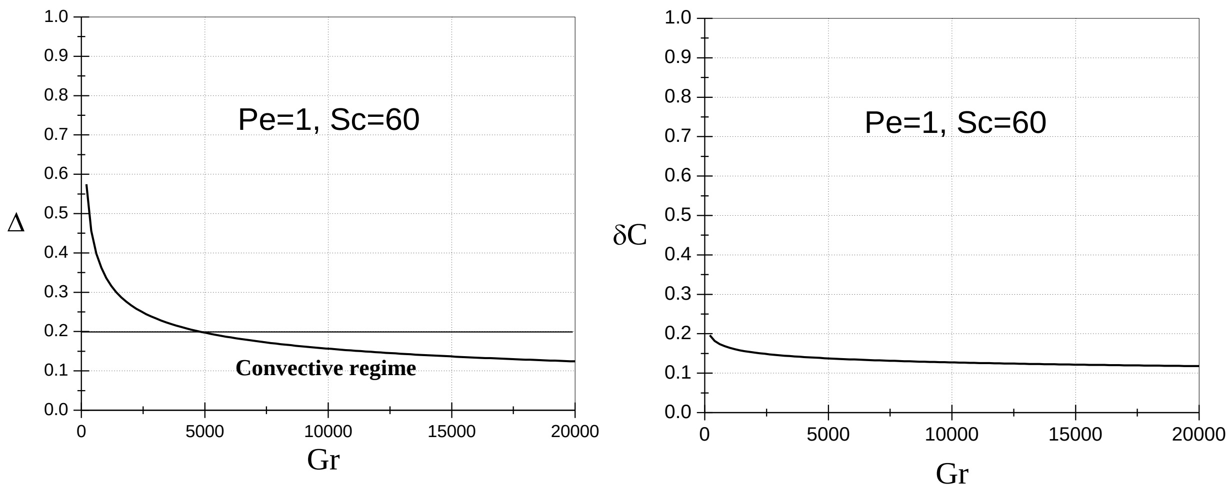


Fig.2 a) Convecto-diffusive parameter Δ as function of the Gr number; b) Radial segregation δC as function of the Gr number. Growth rate $V = 2\mu m/s$.

In conclusion, the parametric study shows that the growth rate has a significant influence on the growth of crystals under convective regime. It is found that at the same intensity of convection, slow growth rates are favorable for obtaining a full convective regime in the melt.

In the following, the influence of the diffusion coefficient on the convecto-diffusive parameter and the radial segregation is analyzed. For the precedent cases the coefficient diffusion was $D = 10^{-8} m^2/s$.

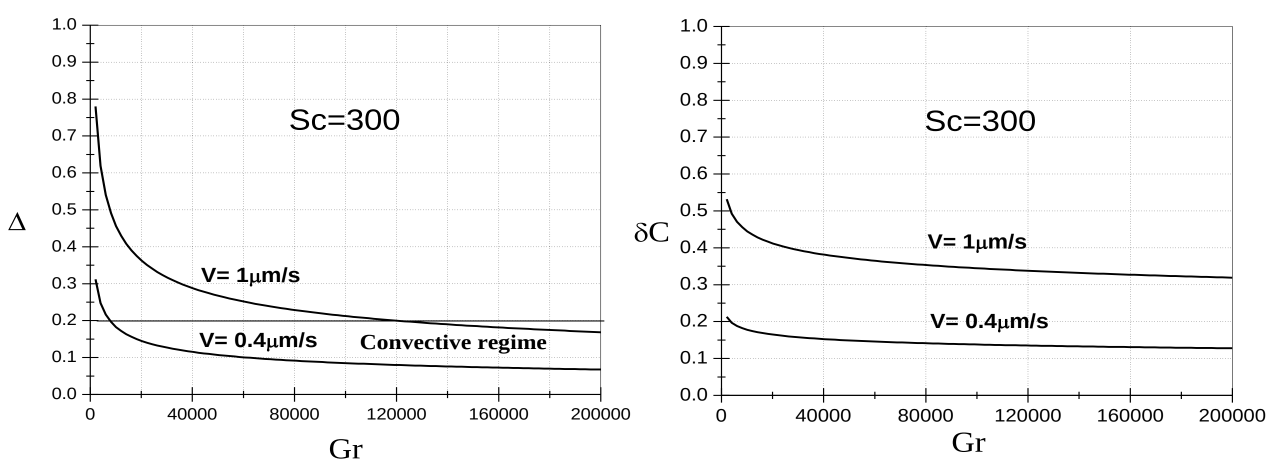
In the Fig.3, the parameter Δ and the radial segregation are represented as function of the Grashof number for a diffusion coefficient $D = 0.5 \cdot 10^{-8} m^2/s$ at the growth rate $V = 1\mu m/s$. It is found a convective regime for $Gr > 5000$ and radial segregations $\delta C < 0.15$, so pull rates of $1 \mu m / s$ are favorable for growing these crystals.



a) b)

Fig.3 a) Convecto-diffusive parameter as function of the Gr number; b) Radial segregation as function of the Gr number. Diffusion coefficient $D = 0.5 \cdot 10^{-8} m^2 / s$, growth rate $V = 1 \mu m / s$.

When the diffusion coefficient has small values ($D = 10^{-9} m^2 / s$), the convective regime is obtained at huge values of the Grashof number $Gr > 120000$ and the radial segregation increases (see Fig.4). In this case, a convective regime at reasonable levels of convection can be obtained by reducing the growth rate to $V = 0.4 \mu m / s$.



a) b)

Fig.4 a) Convecto-diffusive parameter as function of the Gr number; b) Radial segregation as function of the Gr number. Diffusion coefficient $D = 10^{-9} m^2 / s$, growth rates $V = 1 \mu m / s$ and $V = 0.4 \mu m / s$.

Conclusions

The parametric analysis performed by varying the growth rate and the diffusion coefficient can predict the conditions for obtaining a convective regime in the melt. It is found that the growth rate has a significant influence on the convecto-diffusive transport conditions in the melt.

As the growth rate increases, a convective regime characterized by small radial segregations is obtained at higher values of the Grashof number. When $Gr > 10000$, the flow becomes intense and has influence on the thermal field and the shape of the solid-liquid interface. This has a negative influence on the growth process and the solute distribution, so very fast pulling rates should be avoided. The parametric study shows optimal values of the growth rate of $V = 1\mu m/s$ for the analyzed case.

The influence of the diffusion coefficient is also very important. For materials with very small values of the diffusion coefficient $D = 10^{-9} m^2/s$, a convective regime can be obtained at huge values of the Grashof number ($Gr > 120000$) at $V = 1\mu m/s$. A full mixing regime at reasonable levels of convection can be obtained by reducing the growth rate ($V = 0.4\mu m/s$). In this case $Gr > 7000$, and the radial segregation decreases $\delta C = 0.15$.

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