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Combined-convection segregation coefficient and related Nusselt numbers

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ABSTRACT

In Czochralski growth, forced convection is always accompanied by natural convection. Hence, an effective segregation coefficient, which accounts for both forced and buoyancy-induced convection, was recently proposed. This combined-convection coefficient is presented here in dimensionless form, k_{CC} (Nu_{FC}, Nu_{NC}, Pe), where Nu_{FC} and Nu_{NC} are Nusselt numbers for forced and natural convection and Pe is Peclet number for convection due to freezing. Several Nu-number correlations, relevant for Czochralski growth, are presented.

The seminal diffusion coefficients of dopants in molten Si, reported by Kodera, are revisited. Kodera's calculations were done using the BPS model, which does not account for buoyancy-induced flow. In a part of Kodera's experiments, natural convection was significant, and has "inflated" Kodera's coefficients D [cm^2/s].

The k_{CC} correlation is applied to Kodera's data to provide more precise values of D. The impact of buoyancy-induced flow on CZ segregation is demonstrated.

1. Introduction

The equilibrium segregation coefficient, $k_0 \equiv C_S/C_0$ is a fundamental property of any solid—liquid binary system, and appears in a great number of mathematical expressions involving redistribution of the solute [1]. It relates the solute concentration in the solid C_S, to the interfacial solute concentration C_0, in the melt. During solidification at a finite freezing rate f, the solute rejected at the solid—liquid interface, forms a solute boundary layer, as it is carried away by convection. In the presence of convection, one uses the effective segregation coefficient $k_{eff} \equiv C_S/C_L$, to relate C_S to concentration in the bulk liquid phase, C_L.

1.1. Forced convection models of k_{eff}, based on film-thickness

In the seminal 1953 paper, Burton, Prim and Slichter [2] introduced the BPS model or theory, where the level of melt convection is quantified by a fictive "diffusion boundary layer" of thickness δ [1]. The BPS segregation coefficient k_{BPS} is an effective segregation coefficient. It is a useful parameter for crystal growth, because it correlates the composition of the pulled rotating crystal, to composition of the bulk melt [1].

The BPS model is comprised of two equations. The first equation relates k_{BPS} to the thickness of the fictive static solute layer δ_{static},

$$k_{BPS} = \frac{C_S}{C_L} = \frac{k_0}{1 + (1 - k_0)\exp\left(-\frac{f\delta_{static}}{D}\right)} \qquad \text{BPS} \tag{1}$$

where D is diffusion coefficient. Solute transfer through δ_{static}, is by diffusion only (no fluid flow). The second BPS equation, is used to calculate the solute-layer thickness, controlled by the flow induced by a rotating disk,

$$\delta_{FC} = 1.61 D^{1/3} \nu^{1/6} \omega^{-1/2} = 1.61\left(\frac{\nu}{\omega}\right)^{1/2} Sc^{-1/3} \qquad \text{BPS} \tag{2}$$

where $Sc = \nu/D$ is Schmidt number, ω is rotation rate, and ν is kinematic viscosity. The subscript FC is used to highlight the fact that melt is stirred by forced convection. The assumption $\delta_{static} = \delta_{FC}$ remains a weak spot of the BPS theory.

Equation BPS (2) is an outcome of the Levich's asymptotic ($Sc \to \infty$) solution for the mass flux j, in laminar flow, driven by an infinite rotating disk [3],

$$j = -D\frac{dC}{dx}\bigg|_{x=0} = D\frac{\Delta C}{\delta_{FC}} = 0.62 D\left(\frac{\nu}{\omega}\right)^{1/2} Sc^{1/3} \Delta C \tag{3}$$

In 1963, Kodera employed the BPS model to calculate the diffusion coefficients of dopants in silicon melts [4]. These diffusion coefficients are used in most handbooks, although the soundness of the BPS model has been challenged [5–8]. Several alternative models for k_{eff}, all based on film thickness δ, were proposed [5–8].

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In summary, the effective segregation coefficients based on δ_{FC}, accounts for laminar disk-driven convection, and not for buoyancy-induced. Furthermore, δ_{FC} does not account the freezing rate f, while it is evident that $\delta \searrow$ for $f \nearrow$.

2. Combined (or mixed) convection coefficient

In the presence of Earth gravity, buoyancy-induced convection is unavoidable; forced convection is always accompanied by natural convection.

Mixed convection is combined forced and buoyancy-induced convection. In the heat and mass transfer literature [9,10], it is well established that parameter Gr/Re^2 scales the relative importance of natural convection relative to forced convection. The Grashof and Reynolds number are,

$$Gr = \frac{g(\Delta\rho/\rho)H^3}{\nu^2} \tag{4}$$

$$Re = \frac{\omega R^2}{\nu} \tag{5}$$

where $g = 981$ cm/s^2 is gravitational acceleration, H is melt height and R is disk radius. For

$$\left.\begin{array}{ll} a)\ Gr/Re^2 < 0.1, & \text{natural convection is negligible.} \\ b)\ Gr/Re^2 > 10, & \text{forced convection is negligible.} \\ c)\ 0.1 < Gr/Re^2 < 10, & \text{neither is negligible.} \end{array}\right\} \tag{6}$$

Convection fluxes are quantified using convection coefficients h [cm/s] [9,11],

$$j = -D\frac{dC}{dx}\bigg|_{x=0} = h\Delta C \tag{7}$$

or dimensionless Nusselt[1] numbers,

$$Nu \equiv \frac{h\,R}{D}$$

where R is characteristic length, e.g. crystal radius.

2.1. Combined-convection segregation coefficient, k_{CC} (Nu_{mix}, Pe)

Segregation in CZ growth is controlled by combined: (a) forced convection due to crystal rotation; (b) buoyancy-induced natural convection and (c) forced convection due to freezing. Such combined convection is accounted for in the recently proposed formula [11], which is given here as a function of dimensionless parameters,

$$k_{CC}(Nu_{mix}, Pe) = \frac{C_S}{C_L} = \frac{k_0}{1-(1-k_0)Pe/(Nu_{mix}^n + Pe^n)^{1/n}} \tag{8}$$

where,

$$Pe \equiv \frac{f\,R}{D}$$

is the Peclet number, accounting for convection due to freezing. Nu_{mix} accounts for mixed convection [9,11],

$$Nu_{mix}^m = Nu_{FC}^m + Nu_{NC}^m \tag{9}$$

which is comprised of forced (Nu_{FC}) and natural (Nu_{NC}). m is 3–4 [9]. Eqs. (8) and (9) are combined in a single correlation for k_{CC},

$$k_{CC} = \frac{k_0}{1-(1-k_0)Pe/Nu_{CC}} \tag{10}$$

where the combined-convection Nusselt number is

$$Nu_{CC}^n = (Nu_{FC}{}^m + Nu_{NC}{}^m)^{n/m} + Pe^n$$

Note that Nu_{CC} depends on the freezing rate f, while δ_{FC}, does not. Further advantages of the combined-convection coefficient k_{CC} compared to k_{BPS} are

(a) buoyancy driven convection, unavoidable on Earth, is accounted for;
(b) numerous correlations for Nu_{FC} and Nu_{NC} are available for laminar or turbulent flow, and finite Sc numbers.

Thus, Nu_{CC} is a “global” convection coefficient, which accounts for combined: (i) forced convection due to crystal rotation; (ii) buoyancy-induced natural convection and (iii) forced convection due to freezing.

Next we present a number of Nu_{FC} correlations, relevant for CZ growth.

2.2. Forced disk-driven convection

Convection driven by rotating disks has been considered in much detail in the monograph “Hydrodynamic Resistance and Heat Loss of Rotating Solids” by Dorfman (1963) [13]. For forced convection, the heat transfer correlations (HT) are converted to mass transfer, using the following transformation [9–11,14],

$$Nu_{HT} = \frac{h_{HT}R}{\lambda} \rightarrow Nu = \frac{hR}{D} \tag{11}$$

$$\mathrm{Pr} \rightarrow Sc$$

where h_{HT} is convection heat transfer coefficient and λ is thermal conductivity. The recent monograph by Shevchuk (2009) gives a review of correlations for convective heat and mass transfer [14]. Next we list these applicable to the Czochralski (CZ) process.

2.2.1. Nu for laminar convection and $1 < Sc < \infty$

For laminar flow driven by a rotating disk, Nu is not a function of the radial distance r. Thus, the average Nu number is equal to the local number. The general form of a correlation for the mass transfer Nusslet number in laminar flow is [14,15]:

$$Nu_{FC} = \frac{hR}{D} = CRe^{1/2}Sc^{1/3} \tag{12}$$

where C is a coefficient. Eq. (3) combined with definition (7) yields,

$$h_{FC}^{Sc\rightarrow\infty} = 0.62D\left(\frac{\omega}{\nu}\right)^{1/2}Sc^{1/3}$$

and,

$$Nu_{FC}^{Sc\rightarrow\infty} = 0.62\left(\frac{\omega R^2}{\nu}\right)^{1/2}Sc^{1/3} = 0.62Re^{1/2}Sc^{1/3} \tag{13}$$

Shevchuk's book, in p. 36 provides the exact values of $Nu_{FC}/Re^{1/2}$ for $1 < Sc < \infty$ [14]. The exact values are plotted in Fig. 1 along with Eq. (13) which gives 17.2% error for $Sc = 10$. We recently proposed [11],

$$Nu_{FC}^{5<Sc<100} = 0.485Re^{1/2}Sc^{0.373} \tag{14}$$

which gives less than 1% error in the $10 < Sc < 130$ range, and less than 4% error for $5 < Sc < 500$, range see Fig. 1. The correlation presented in Newman's book [16], also shown in Fig. 1, gives less than 4% error for $2 < Sc < \infty$,

$$Nu_{FC}^{2<Sc<\infty} = 0.62045Re^{1/2}Sc^{1/3}/(1 + 0.2980Sc^{-1/3} + 0.14514Sc^{-2/3}) \tag{15}$$

[1] The mass-transfer Nusselt number (Nu), also known as the Sherwood number (Sh).