Coupling of conductive, convective and radiative heat transfer in Czochralski crystal growth process

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Abstract

This paper studies the conjugate problems of fluid flow and energy transport (involving conduction, convection and radiation heat transfer) within a material changing its phase. The analysis focuses on the Czochralski crystal growth process. The solidifying material is treated as a pure substance with constant material properties. The solution of the resulting 3-D, axisymmetric, non-linear problem is obtained iteratively using the commercial CFD package Fluent. The algorithm employed here treats each subdomain of the system separately, i.e. the liquid and solid phases of the solidified material, as well as the inertial gas surrounding both phases.

Results of a test case shows the velocity field and temperature distribution within a simple system employed for the growth of a single silicon crystal.
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1. Introduction

The Czochralski crystal growth process is nowadays commonly used to manufacture high quality crystals, e.g. silicon bulk crystals. In this process, a cylindrical crystal grows due to the phase change phenomena occurring in the melt, and is then vertically pulled out of the system. The melt is placed in a cylindrical crucible, located in a furnace, and is heated above the melting temperature by a resistive or inductive electrical heater. The quality of the growing crystal depends on various quantities and phenomena, but the principal ones generally are heat transfer processes and environmental thermal parameters. Thermal parameters certainly influence the heat and mass transfer in the system, interfacial phenomena, as well as the transport of dopants. Therefore, modelling of the heat and mass transfer processes within the system

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is essential for better understanding and controlling the crystal growth process.

There are a number of reasons why the mathematical modelling of the heat transfer problem in these kind of processes is considered to be a very challenging task. The main ones area as follows:

- The mathematical representation of the problem, which also involves the motion of a liquid phase or gas surrounding the solidifying material, requires the solution of a coupled system of continuity, momentum and energy equations;
- The position of the phase change boundary, where internal heat generation takes place as a result of solidification, is unknown and has to be determined as part of the solution;
- Phase change systems usually work at elevated temperatures. This means that thermal radiation may play a crucial role, not only in terms of boundary phenomena, but also within the solidifying or melted medium, if the medium is semitransparent (e.g. some crystals, glass, etc.).

The Czochralski process involves all transport phenomena encountered in solid–liquid phase change systems, including the conjugate conduction–convection–radiation heat transfer. Therefore, controlling Czochralski processes is limited by the degree to which these complex phenomena are properly understood and modelled.

The first published works on the mathematical modelling of the Czochralski process have neglected convection in the melt, radiation in the system and/or assumed a planar interface between melt and crystal. Only recently have these problems received appropriate attention, e.g. [1–9]. Nowak and co-workers have also solved the problem [10,11,14] as a conjugate conduction–convection–radiation heat transfer process resulting from continuous solidification of a semitransparent medium. The problem has been analyzed through the coupling of two computer codes:

- the commercial CFD package Fluent [12],
- an in-house boundary element method (BEM) code which enables radiation to be considered [13].

The CFD package Fluent was employed in [10,11] to solve a conjugate system of equations, for the liquid and solid phases, consisting of the continuity equation, the momentum equations (in the liquid phase only) and the energy equation. It is important to notice that the latter equation contains volumetric heat sources resulting from thermal radiation occurring within the system. These sources were calculated by the in-house code using typical BEM algorithms. Thus, by using the two codes in succession, the velocity field in the liquid phase and the resulting temperatures and heat fluxes have been calculated. However, some difficulties were experienced with convergence in the *Fluent* code, particularly in the liquid phase for higher values of the thermal expansion coefficient. To avoid this drawback, a new approach for solving the problem is proposed in this paper. The new formulation is based on the assumption that the heat transfer and fluid flow problems can be solved by the Fluent package separately for each phase, and the solutions thus obtained are then coupled along the interfaces by taking into account appropriate interfacial boundary conditions. These conditions can only be satisfied through an iteration process, which starts with an initial guess and is gradually adjusted until convergence.

Generally, there are two kinds of interfaces within the system. The interfaces between an inert gas surrounding the solidifying material (e.g. argon) require adjusting both temperatures and heat fluxes; however, their location is knwon in advance. In contrast to that, the temperature along the phase change front is known but both the heat flux and the phase change front location have to be determined.

The main aim of this paper is to examine the methodology of coupling the solutions obtained separately by the Fluent code for each subdomain, i.e. liquid and solid phases of the solidifying material and gas. Coupling these solutions requires a new in-house code which iteratively adjusts the continuity of temperature and heat fluxes along the gas–liquid and gas–solid interfaces, as well as the Stefan boundary conditions on the phase change interface. At this stage of analysis, thermal volume radiation within the crystal is neglected,

although surface thermal radiation is still considered.

2. Problem formulation

To discuss the fundamentals of the proposed methodology, a 3-D axisymmetric heat transfer problem of Czochralski crystal growth is analyzed. Due to the symmetry of the problem, only a cross-section of the domain is schematically shown in Fig. 1. The domain consists of three subdomains, i.e. liquid and solid where the position of the interface EH has to be determined, and the gas subdomain.

A liquid material is contained in the crucible, which is heated above the liquidus temperature. The liquid then solidifies due to heat losses from the system, and a crystal is pulled out at a known, constant rate. The solidifying material is treated as a pure substance. Thus, its phase change takes place at a fixed temperature. As previously mentioned, volume thermal radiation is neglected in all subdomains.

Line CDK in Fig. 1 represents the side wall of the crucible, which is heated by a special heater, while the dashed line KJF represents a cover minimizing heat losses from the system. In this paper, it is assumed that both these surfaces are maintained at a temperature typical of the relevant industrial application. It should also be noted that the temperature of these surfaces are important parameters for controlling the process.

Since the surface KJF is non-transparent, all energy arriving there by radiation is partially absorbed and partially reflected (according to the emissivity of the surface), see e.g. [13].

2.1. Governing equations

The boundary-value problem is formulated in a coordinate system attached to the crucible. This leads to the following system of conjugate steady-state heat transfer equations:

$$\nabla^2 T(\mathbf{r}) - \frac{1}{a}\vec{v}\cdot\overrightarrow{\nabla T} = 0 \tag{1}$$

$$\vec{\nabla}\cdot\vec{v} = 0 \tag{2}$$

$$\varrho(\vec{v}\cdot\vec{\nabla})\vec{v} = -\overrightarrow{\nabla p} + \eta\nabla^2\vec{v} + \varrho\vec{g}\beta(T - T_\infty) \tag{3}$$

where T is the temperature at point $\mathbf{r}$, a stands for the thermal diffusivity, λ denotes the heat conductivity while $\vec{v}$ represents the velocity vector. Density is represented by ϱ, p stands for the pressure, η is viscosity while β and $\vec{g}$ are the coefficient of thermal expansion and the gravitational vector, respectively. Buoyancy effects are modelled through the Boussinesq approximation.

The above system of equations is valid for both the liquid and gas subdomains. The energy Eq. (1) also governs the heat transfer in the solid, but in this case $\vec{v} = \vec{v}_x$ is the known, constant pulling velocity v_{GF}.

2.2. Boundary conditions

External boundary conditions are fairly standard, i.e. of the Dirichlet, Neumann and/or Robin types, and as such will not be discussed herewith. It should only be remembered that most of these boundary conditions also account for radiative heat fluxes.

The conditions along the phase change front (surface EH) and the interfaces DE and EF are obviously more interesting. The condition along the phase change front involves continuity of temperature but a jump in the heat flux across this interface. Additionally, the common temperature on both sides of the interface has to be equal to the melting temperature, i.e.

$$T_L(\mathbf{r}) = T_S(\mathbf{r}) = T_{ph} \tag{4}$$

Fig. 1. Domain under consideration.

$$-\lambda_L \frac{\partial T_L}{\partial n_{ph}} + \lambda_S \frac{\partial T_S}{\partial n_{ph}} = -l_{ph}\varrho_S\left(\vec{v}\cdot\vec{n}_{ph}\right) \qquad (5)$$

where T_{ph} is the phase change temperature, l_{ph} stands for the latent heat while $\vec{n}_{ph}$ represents the vector normal to the phase change surface. The subscripts L and S refer to liquid and solid phases, respectively.

Simultaneously, the following no-slip boundary condition holds for the melt velocity:

$$\vec{v}_L = \vec{v}_x \qquad (6)$$

The conditions along the interfaces DE and EF generally impose continuity, but modified by radiative heat fluxes, arriving from the gas phase, at the liquid or solid surface, i.e.

- *along face DE*

$$T_L(\mathbf{r}) = T_G(\mathbf{r}) \qquad (7)$$

$$\lambda_L \frac{\partial T_L}{\partial n} = \lambda_G \frac{\partial T_G}{\partial n} + q_{rL} \qquad (8)$$

where q_{rL} is a radiative heat flux arriving at the liquid surface, at a fixed interface node.

- *along face EF*

$$T_S(\mathbf{r}) = T_G(\mathbf{r}) \qquad (9)$$

$$\lambda_S \frac{\partial T_S}{\partial n} = \lambda_G \frac{\partial T_G}{\partial n} + q_{rS} \qquad (10)$$

where q_{rS} is a radiative heat flux arriving at the solid surface, at a fixed interface node.

3. Solution procedure

The commercial CFD package Fluent has been employed to solve the coupled diffusion–convection–radiation boundary-value problem formulated in the previous section. This package is based on the finite volume method, and requires all relevant boundary conditions to be defined. As previously mentioned, in the proposed approach Fluent is used separately for each selected subdomain, i.e. the liquid, solid and gas subdomains. Therefore, there is a need for coupling the solu-

tions related to these subdomains within external iterative loops.

The Stefan boundary conditions (4) and (5) are used to couple the solutions for the liquid and solid phases, as well as to correct the position of the interface. The procedure consists of the following steps:

(1) assume an initial position of the interface EH;
(2) define the remaining boundaries of the solid, liquid and gas subdomains, as well as their discretizations (which are kept constant throughout the analysis);
(3) solve the boundary-value problem for the liquid subdomain, assuming conditions (7) and (8) along the interface DE, and a constant temperature equal to the phase change temperature T_{ph} along the trial interface EH;
(4) calculate the heat flux distribution $q_{Sn_{ph}}(\mathbf{r}) = -\lambda_L(\partial T_L/\partial n_{ph})$ along the solid side of the trial interface EH, employing the boundary condition (5), i.e.

$$q_{Sn_{ph}}(\mathbf{r}) = q_{Ln_{ph}}(\mathbf{r}) + l_{ph}\varrho_S\left(\vec{v}\cdot\vec{n}_{ph}\right) \qquad (11)$$

(5) solve the boundary-value problem for the solid subdomain, assuming conditions (9) and (10) along the interface DE, and the above-calculated heat flux densities along the trial interface EH;
(6) verify the calculated temperature distribution along the interface, and terminate iterative loop if the differences between nodal temperature values on the interface and the phase change temperature are sufficiently small;
(7) if not, define a new interface position and return to step 2.

A new interface position is obtained by relocating nodes belonging to the interface EH. These nodes are displaced along the x direction only. At the $m+1$ iteration step, the new coordinate $x_{i,m+1}$ of the nodal point i located on the interface is calculated by using the following heuristic formula:

$$x_{i,m+1} = x_{i,m} - \omega\left(T_{i,m} - T_{ph}\right)\frac{x_{i,m} - x_{i+1,m}}{T_{i,m} - T_{i+1,m}} \qquad (12)$$

where subscripts i and $i+1$ are related to two neighbouring nodes (the actual one and the first off the interface) having the same y coordinate. The symbol ω denotes a relaxation coefficient used to control convergence. In the present calculations, the value of ω ranges from 0.2 to 0.5.

4. Numerical example

To demonstrate the main features of the proposed algorithms, a numerical example of Czochralski crystal growth process has been solved. The domain under consideration consists of silicon [2] (treated here as non-transparent material) and a hot gas (argon), as schematically shown in Fig. 1.

The thermophysical data and operating parameters can be found in [2] and [10]. It should be noted that the density of the melt depends on the coefficient of thermal expansion β and the temperature, as given by the Boussinesq approximation. Thus, the density ϱ of melt refers only to the melting point temperature.

The discretization adopted assumes a division of the melt into 50×60 elements, and a divison of the solid and gas subdomains into 30×80 elements.

The value of the absorption coefficient, essential for the radiation analysis, was assumed to be equal to 0.85 for the crucible (i.e. surface DK), as well as for the surfaces KJ and JF. A value of 0.7 was assumed for the emissivity of the crystal surface (EF), while a value of 0.3 was assumed for the melt surface (DE).

A typical temperature distribution is shown in Fig. 2, for the domain depicted in Fig. 1. In this figure, the 1683 K isotherm shows the position of the phase change front. The numerical solution was obtained in 20 iterations, starting from a horizontal position of the interface EH.

Velocity profiles within all subdomains are shown in Fig. 3. Because different scales for the velocity arrows are used in each subdomain, it is important to remember that the maximum velocity found in the liquid phase is equal to 0.00365 m/s, while in the gas phase this quantity is equal to 0.165 m/s. Velocities in the solid are all equal to 1.39×10^{-5} m/s (the pulling rate).
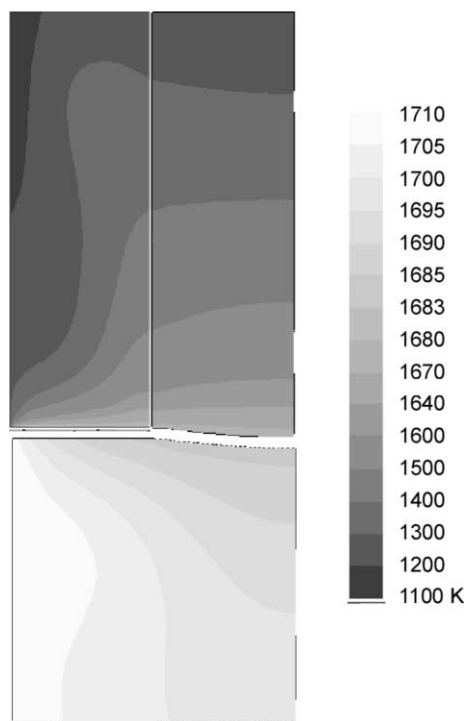

Fig. 2. Temperature distribution within cross-section of the considered domain.

The results shown in Figs. 2 and 3 are natural extensions of the authors' previous works [10,11,14]. The numerical results obtained here are also quite similar to results presented by other authors, who performed even more rigorous analysis of the crystal growth process but using different numerical methods, e.g. [8,9]. Although it is not possible to quantitatively compare such results, a good qualitative agreement is evident.

5. Conclusions

It has been demonstrated how the commercial CFD software Fluent can be used to model the steady-state conjugate problem involving heat conduction, convection and surface thermal radiation. The present simulation applied Fluent separately for each subdomain (i.e. solid, liquid and gas phases), and an additional in-house code and an additional iterative loop were thus required.
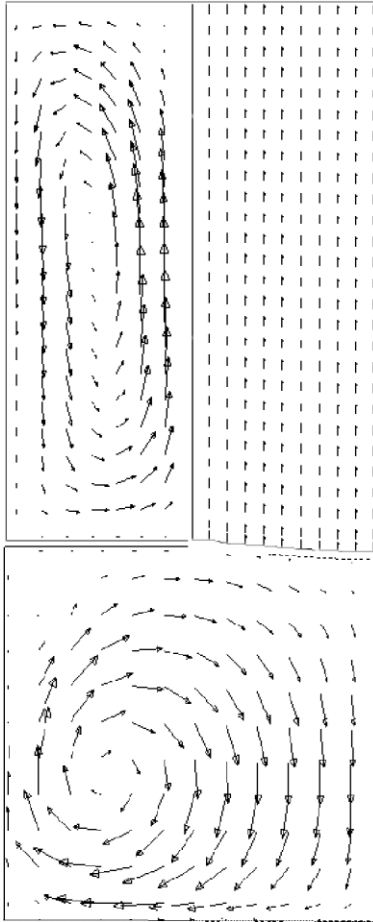
Fig. 3. Velocity profiles within cross-section of the considered domain.

Nevertheless, the proposed algorithm is quite straightforward and converges fairly fast. Such approach takes advantages of many valuable techniques already available in Fluent, including extensive pre- and post-processing facilities.

The computations proved the importance of thermal radiation analysis. The coupling of radiation with other heat transfer modes discussed in this paper showed that the mathematical model and solution procedures are compatible and robust. Only five external iterations were required for the final solution of the numerical example presented here. Such solution offers detailed information about temperatures and heat fluxes, as well as velocity profiles and pressure distribution, within the system.

Of course, many improvements to the proposed algorithm are possible, and will be the subject of further research. Nevertheless, even in its current state, the approach described in this paper can already be used to control the Czochralski crystal growth process.

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