Advanced inverse techniques for the design of directional solidification processes

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Abstract

New advanced methodologies for thermal process design can be developed using inverse problem theory. Such techniques will be presented here for the design of directional solidification processes. In particular, we will address the problem of designing the mold cooling/heating conditions such that stable solidification growth is obtained with control of the growth rate at which solidification occurs as well as of the temperature field near the solid/melt interface. Changes in growth rate and thermal gradient at the freezing interface are the driving forces dictating the formation of specific microstructures.

The methods of choice for the solution of the above problems are functional optimization methods. We will present newly developed continuum inverse formulations based on the adjoint method for binary alloy systems with coupled heat, mass and melt flow transport mechanisms. The unknown controls (e.g. the mold heat flux) are assumed to be functions in the L_2 space and exact calculation of the gradient of the objective function has been performed. An example is presented for the design of stable directional solidification with a desired growth.

Various design, identification and control problems take the form of an *inverse problem* in which incomplete conditions are available in part of the boundary, whereas overspecified boundary conditions have been supplied in another part of the boundary or inside the domain. Such design conduction based solidification problems are given in [1]-[4].

In our recent work [5]–[6], we derived a functional optimization formulation and continuum adjoint equations for inverse natural convection problems. This paper will generalize the analysis of [5] and [6] to the inverse design of directional solidification processes of dilute binary alloys. A design problem will be defined in order to obtain a *desired stable growth* for a binary alloy system. The design is achieved with proper selection of the heating/cooling conditions on the mold walls. An example problem is considered for the solidification of a NH_4Cl water solution in a rectangular mold. Finally, potential extensions of the present analysis to other solidification design problems are discussed.

A reference design for desired freezing front motion

Let us consider the directional solidification process of a dilute binary alloy (Fig. 1(a)). At time $t = 0^+$, a cooling heat flux is applied at the side Γ_{os} of the mold wall and solidification starts. We assume that the solid/liquid interface is a sharp one.

Let us introduce a reference design problem (see Fig. 1(b)) with $q_{ol} = 0$ and a *flat* interface throughout the process of solidification. The inverse *reference design problem* is stated as follows: *Find the appropriate heat flux* $q_{os}(y,t)$ *at* Γ_{os} *such that the freezing interface remains flat and advances with a spatially uniform velocity* $\boldsymbol{v}_f(t)$. The major objective is here the elimination of the effects of double diffusive convection on the freezing interface morphology. We can separate this problem into two subproblems and solve them sequentially, a quasi-direct problem in the liquid domain $\Omega_\ell(t)$ and an inverse heat conduction problem in the solid domain $\Omega_s(t)$.

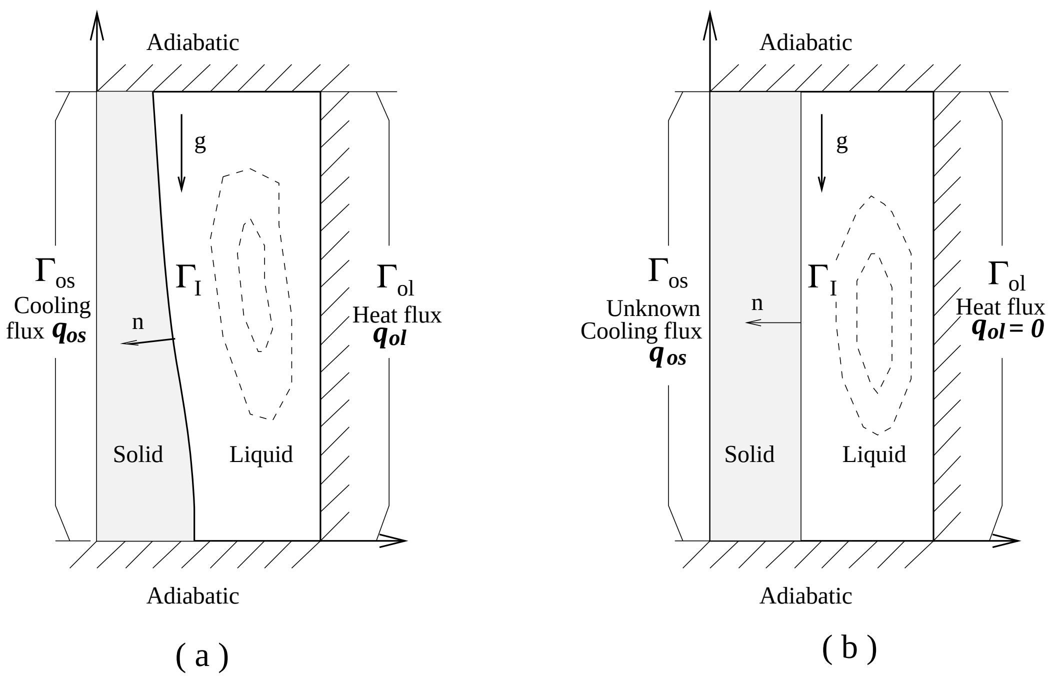
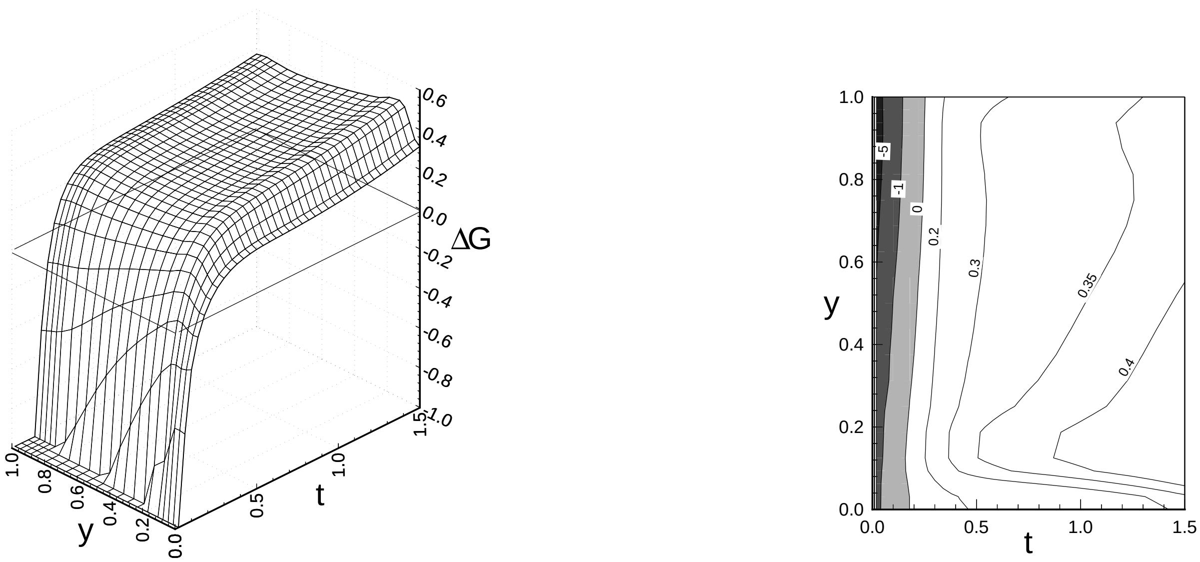


Figure 1: (a) The direct solidification problem with heat flux q_{os} (cooling) on Γ_{os} and heat flux q_{ol} on $\Gamma_{o\ell}$ (b) The *reference inverse design problem* to achieve a flat solid-liquid interface growth with a uniform desired velocity ($q_{ol} = 0$).

Consider the solidification of a NH_4Cl water solution (1.5% weight concentration) with initial overheating of $T_{ini} - T_o = 20°C$, in a rectangular cavity (dimensionless height $h = 1$ and width $w = 0.5$, Fig. 1(b)). The design objective is to realize a uniform growth of $\boldsymbol{v}_f = 0.2$. The thermophysical properties are shown in Table 1. Solidification starts off at

Prandtl number	Pr	$\frac{\nu}{\alpha_\ell}$	9.025
Lewis Number	Le	$\frac{\alpha_\ell}{D_\ell}$	27.845
Partition ratio	k	c_s/c_ℓ on Γ_I	0.3
Relative initial overheat	γ	$(T_i - T_o)/(mc_o) + 1$	18.152
Thermal Rayleigh number	Ra_T	$[\lvert\boldsymbol{g}\rvert\beta_T(T_i - T_{ref})l^3]/(\nu\alpha_\ell)$	2.0×10^4
Solutal Rayleigh number	Ra_C	$[\lvert\boldsymbol{g}\rvert\beta_C c_o l^3]/(\nu\alpha_\ell)$	1.0×10^4

Table 1: Non-dimensional parameters used for $NH_4Cl - H_2O$.



(a) (b)

Figure 2: Examination of the constitutional stability assumption at the solid-liquid interface for the reference design problem: (a) Difference $\Delta G^{ref} = G^{ref} - G_c^{ref}$; (b) contour lines of ΔG^{ref} in the (y, t) plane.

$t = 0^+$ when the $x = 0$ boundary is dropped to $T = 0$ (dimensionless melting temperature at initial concentration), and runs up to $t_{max} = 1.5$ when 60% of the slab has solidified.

The above stated problem can be solved using the FEM techniques given in [5], [6]. However, we will next show that the calculated solution does not satisfy the a-priori assumption of stable growth. Indeed, let us consider the simplified form of stability expressed in terms of the constitutional undercooling condition [7], $G < G_c$, where G and G_c are the gradients in the normal direction of the dimensionless temperature and concentration fields at the liquid side of the interface, respectively. Note that this form of constitutional undercooling is the dimensionless form of the eq. $G > mG_c$ given in [7]. The dimensionless quantities used here are $\bar{c} = (c_o - c)/(\gamma c_o)$, where $\gamma = 1 + (T_{ini} - T_o)/(mc_o)$, with m as the slope of the liquidus line. Also, the normal unit vector $\boldsymbol{n}$ at the interface boundary points towards the solid phase. Let us examine if the solution is such that $\Delta G = G - G_c < 0$ is satisfied. From Fig. 2(a), we observe that $\Delta G > 0$ some time after solidification started. Thus the stability condition is only satisfied at the early stages of solidification (the shaded region of Fig. 2(b)).

The above mathematical model with the assumption of a sharp freezing interface is thus not physically realistic. Next, we will design a directional solidification process in which the effects of thermo-solutal convection on the interface morphology are controlled such that a desired stable growth is obtained (similar to that shown in Fig. 1(b)).

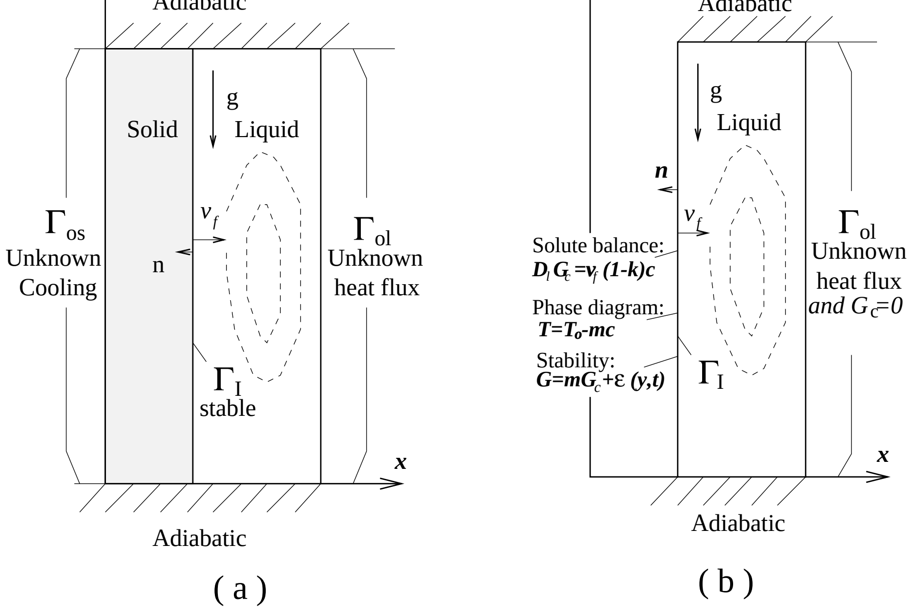


Figure 3: Inverse problem to achieve a sharp interface moving with a desired growth velocity $\boldsymbol{v}_f$: (a) Problem unknowns; (b) Inverse subproblem in the liquid.

Inverse design to achieve a desired stable growth

In order to achieve a desired interface growth, we relax the adiabatic condition q_{ol} at the mold wall $\Gamma_{o\ell}$ in the reference problem of Fig. 1. We pose using Fig. 3(a) the following inverse design problem: *Find the cooling condition at* Γ_{os} *as well as the heat flux condition* $q_{o\ell}(y,t)$ *at* $\Gamma_{o\ell}$*, to achieve a desired growth of the interface (the same as that in the reference problem of Fig. 3(a)) that is ensured to be constitutionally stable.*

Such an inverse problem can be separated into two subproblems, one inverse problem in the solid and another inverse problem in the liquid region. The inverse problem in the solid is an inverse heat conduction problem. The inverse problem in the liquid is defined in Fig. 3(b). We call such an inverse design problem *an inverse design thermal-solutal convection problem*. For this problem, we transform the stability requirement to the following non-dimensional equality form:

$$\frac{\partial T}{\partial n} = \frac{\partial c}{\partial n} + \epsilon(y,t) \qquad (1)$$

where $\epsilon \leq 0$. We will here use the calculated $\Delta G^{ref}(y,t)$ in the reference design problem (Fig. 2(a)) as a basis for choosing ϵ, and make necessary adjustments to ϵ when ΔG becomes positive (i.e. when the interface of the reference design became unstable). In particular, we select the following form of the stability condition:

$$\Delta G(y,t) \equiv \epsilon(y,t) = \begin{cases} \Delta G^{ref}(y,t), & \text{if } \Delta G^{ref}(y,t) < \delta Le\gamma^{-1}(1-k)(\boldsymbol{v}_f(t)\cdot\boldsymbol{n}) \\ \delta Le\gamma^{-1}(1-k)(\boldsymbol{v}_f(t)\cdot\boldsymbol{n}), & \text{if } \Delta G^{ref}(y,t) \geq \delta Le\gamma^{-1}(1-k)(\boldsymbol{v}_f(t)\cdot\boldsymbol{n}) \end{cases} \qquad (2)$$

Here, δ is chosen as a small scalar parameter. As $\delta \to 0$, $\epsilon \to 0$ which means that marginal stability is maintained after $\Delta G^{ref} > 0$. The underlying physics of such a choice is to pursue minimum heating flux input at the liquid boundary mold wall $\Gamma_{o\ell}$, thus minimum cooling at the solid mold wall Γ_{os} and overall a minimum energy consumption that ensures the desired stable growth. Design for an interface that is slightly over-stable will compensate for the numerical error in the implementation of the discretized problem. The specific form of such an over-stable amount can be understood from its equivalent dimensional form:

$$G = -m\left[G_c - \delta\frac{c_o}{D_\ell}(1-k)(\boldsymbol{v}_f(t)\cdot\boldsymbol{n})\right] \qquad (3)$$

fixed $c = c_o$.

The inverse design thermal-solutal convection problem in the liquid (Fig. 3(b)) can be formulated as an optimization problem. With a *guessed* heat flux $q_{o\ell}(\boldsymbol{x},t), (\boldsymbol{x},t) \in \Gamma_{o\ell} \times [0, t_{max}]$, one can define a direct thermal-solutal convection problem on the given domain $\Omega_\ell(t)$. Let us denote its solution for the temperature, concentration and flow fields as $T(\boldsymbol{x},t;q_{o\ell})$, $c(\boldsymbol{x},t;q_{o\ell})$ and $\boldsymbol{u}(\boldsymbol{x},t;q_{o\ell})$. Note that the liquidus equilibrium relation is not used in this *direct problem* definition, thus it is not guaranteed to be satisfied. For an arbitrary $q_{o\ell} \in L_2(\Gamma_{o\ell} \times [0, t_{max}])$, we define a cost functional:

$$S(q_{o\ell}) = \int_0^{t_{max}} \int_{\Gamma_I} \left[T(\boldsymbol{x},t;q_{o\ell}) - c(\boldsymbol{x},t;q_{o\ell})\right]^2 d\Gamma dt, \tag{4}$$

to indicate the discrepancy of the calculated temperature from the concentration-dependent liquidus temperature at the interface. The cost functional can be thought of as a measure of the deviation from thermodynamic equilibrium of the interface.

The inverse problem in the liquid is re-stated in a minimization sense as: *find a quasi-solution* $\bar{q}_{o\ell} \in L_2(\Gamma_{o\ell} \times [0, t_{max}])$ *such that,*

$$S(\bar{q}_{o\ell}) \leq S(q_{o\ell}), \quad \forall \;\; q_{o\ell} \in L_2(\Gamma_{o\ell} \times [0, t_{max}])$$

To perform the optimization procedure that minimizes $S(q_{o\ell})$ in $L_2(\Gamma_o \times [0, t_{max}])$, we will need to define a *continuum sensitivity problem*. This linear problem defines the linear perturbations of the fields $T(\boldsymbol{x},t;q_{o\ell})$, $c(\boldsymbol{x},t;q_{o\ell})$ and $\boldsymbol{u}(\boldsymbol{x},y;q_{o\ell})$, respectively, with respect to variations $\Delta q_{o\ell}(\boldsymbol{x},t)$ of the boundary heat flux $q_{o\ell}$. In order to realize the minimization of $S(q_{o\ell})$, it is also essential to find its gradient (derivative) $S'(q_{o\ell})$ with respect to $q_{o\ell}$. After lengthy manipulations, one can define an appropriate *adjoint problem* such that the gradient of $S(q_{o\ell})$ is given as follows:

$$S'(q_{o\ell}) = \psi(\boldsymbol{x},t;q_{o\ell}), \quad (\boldsymbol{x},t) \in \Gamma_{o\ell} \times [0, t_{max}] \tag{5}$$

The conjugate gradient method can now be used for the minimization of the cost functional $S(q_{o\ell})$. It constructs a sequence: $q^0_{o\ell}$, $q^1_{o\ell}$, $\ldots$, $q^k_{o\ell}, \ldots$, to approach the optimal minimizer $\bar{q}_{o\ell}$. All details of the derivations of the sensitivity and adjoint systems as well as of the gradient calculation can be found in [9].

Numerical results

The inverse algorithm is implemented here for the solidification of $NH_4Cl - H_2O$ as in the reference design problem. The objective here is to find the transient histories of heat fluxes at $x = 0.5$ and at $x = 0$ that result in a stable growth with a desired front velocity given as follows:

$$\boldsymbol{v}_f(t) = \begin{cases} v_o, & 0 \leq t \leq t_{mid} \\ v_o \frac{t_{max} - t}{t_{max} - t_{mid}}, & t_{mid} < t < t_{max} \end{cases} \tag{6}$$

where we select the numerical dimensionless values as $v_o = 0.2$, $t_{mid} = 1.5$ and $t_{max} = 2.0$. An initial guess $q^0_{o\ell}(y,t) = 0$ is made. As a preliminary study of the solution of such inverse problems and in consideration of the computational cost, the Ra and Le numbers used here are lower than the actual numbers under normal laboratory conditions.

For purposes of performing an accuracy study and comparison, both $\delta = 0$ and $\delta = 0.2$ are chosen as the parameter for "over stability" in eq. (2). The $\delta = 0$ case is seeking a heat flux solution that strictly leads to marginal stability. The optimum heat flux solution $\bar{q}_{o\ell}(y,t)$ is shown in Fig. 4. The calculated flux $q^k_{o\ell}(y,t)$ at various CGM iterations is also shown in Fig. 5 (for the case $\delta = 0.2$). A validation procedure is performed to check the accuracy of the interface stability. A quasi-direct problem in the liquid is solved, using the obtained $\bar{q}_{o\ell}(y,t)$ as boundary condition at the vertical liquid side mold wall $x = 0.5$. Using a similar approach as that shown in Fig. 2, G and G_c are computed *a-posteriori* and the contours of ΔG are plotted in Fig. 6. Since the cost functional cannot be reduced to zero exactly, the quasi-direct problem validation will have both positive and negative ΔG

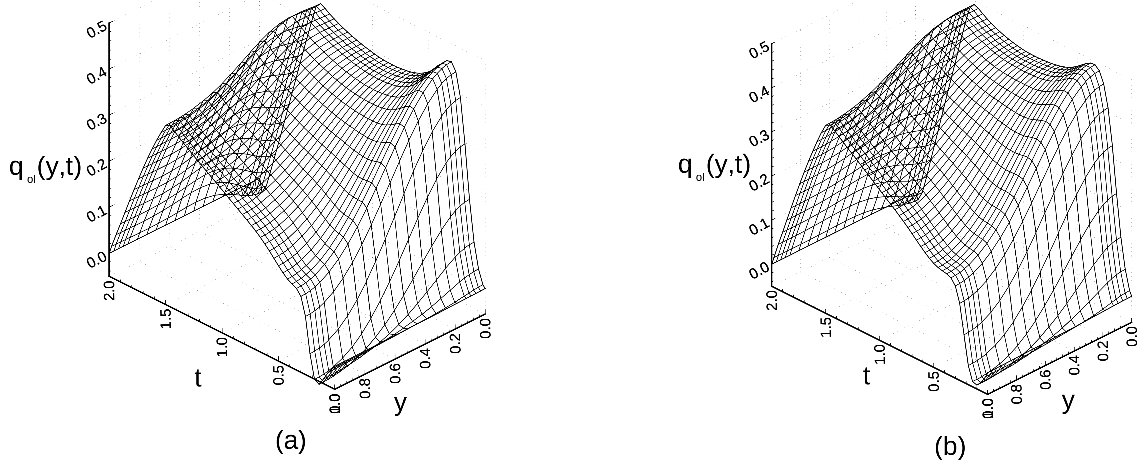


Figure 4: Optimum heat flux $\bar{q}_{o\ell}(y,t)$ for (a) $\delta = 0$ and (b) $\delta = 0.2$. Both results correspond to heating except at the very early stages of solidification.

along the interface if $\delta = 0$ is used in eq. (2) while seeking a marginal stability solution. A non-zero δ solution will reduce ΔG by a certain amount. This amount is enough to overcome the appearance of positive ΔG at all times (except near $t = t_{max}$, see Fig. 6b).

The solution $\bar{q}_{os}(y,t)$ of the inverse heat conduction problem in the solid region is shown in Fig. 7. It is obtained by the adjoint method, using the interface heat flux and temperature from the quasi-direct validation problem with $\delta = 0.2$. Additional non-dimensional parameters involved are $R_k = R_\alpha = 1$ and $Ste = 0.3$. Its combination with $\bar{q}_{o\ell}$ from Fig. 4(b) provides the complete solution that leads to a desired stable interface growth.

Discussion

This paper presented a preliminary algorithm for the mathematical design of directional solidification and crystal growth processes for binary alloy systems. The main difficulty that remains to be addressed for an industrial implementation of such techniques is the development of experimental designs that can actually achieve the proposed optimal boundary heat flux (or temperature) conditions. This remains a technologically challenging and currently open design problem with significant industrial implications. The presented algorithms can however be used to develop experimental designs that provide approximations to the optimal heat fluxes based on the current available experimental cooling/heating mold/furnace control systems.

The present methodology is currently tested for systems with larger Ra and Le numbers. Such studies will extend its applicability to the solidification of semi-conductor materials and to processes with stronger melt convection.

Finally, additional means, such as force convection through electro-magnetic stirring, are necessary approaches together with the boundary heat flux control to design solidification processes. A significant number of mathematical, computational and experimental issues in this direction remain to be addressed.

Acknowledgments

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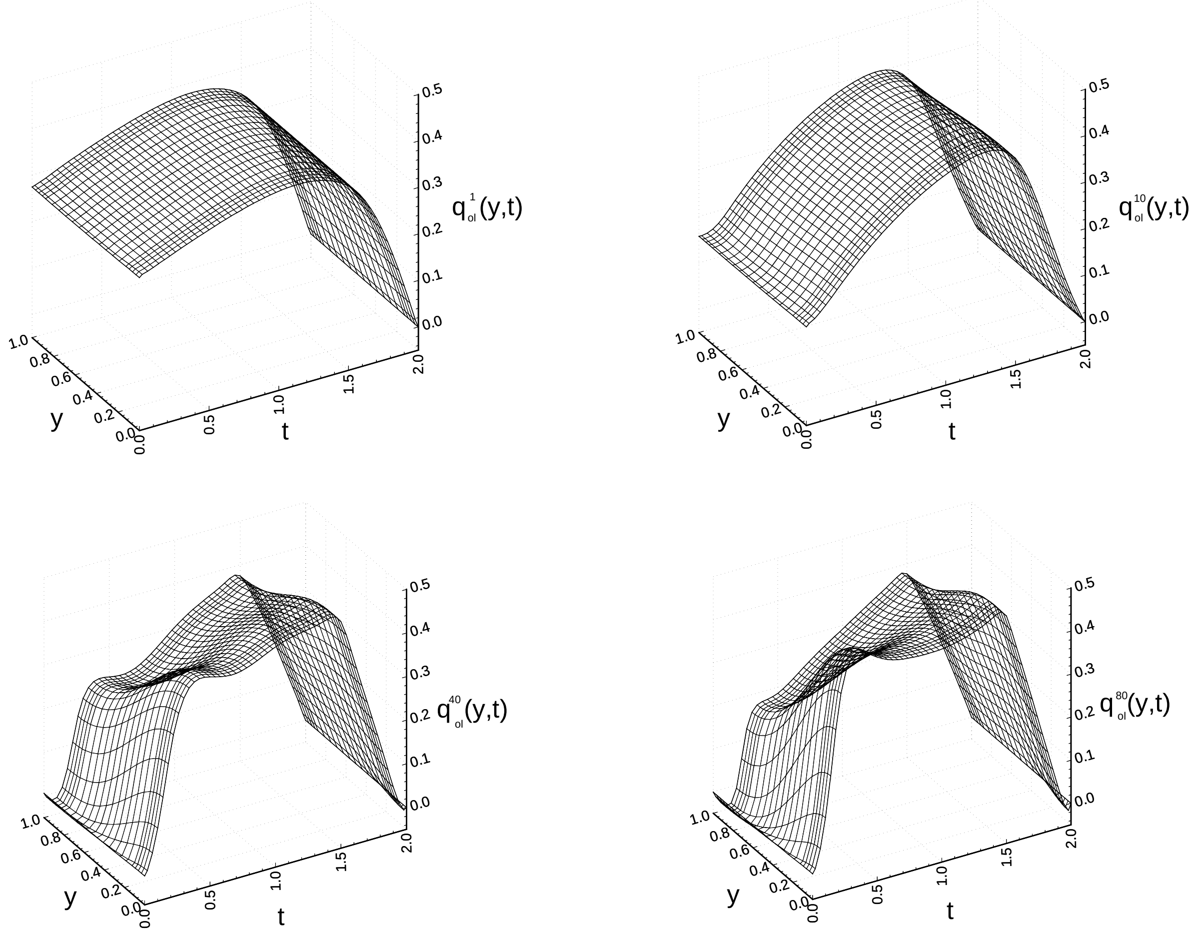

Figure 5: $q_{o\ell}^k(y,t)$ at the intermediate $k = 1,\ 10,\ 40$ and $80'th$ iterations ($\delta = 0.2$). The temporal features of $q_{o\ell}$ are reconstructed at early iterations, whereas accurate reconstruction of the spatial variations requires more iterations.

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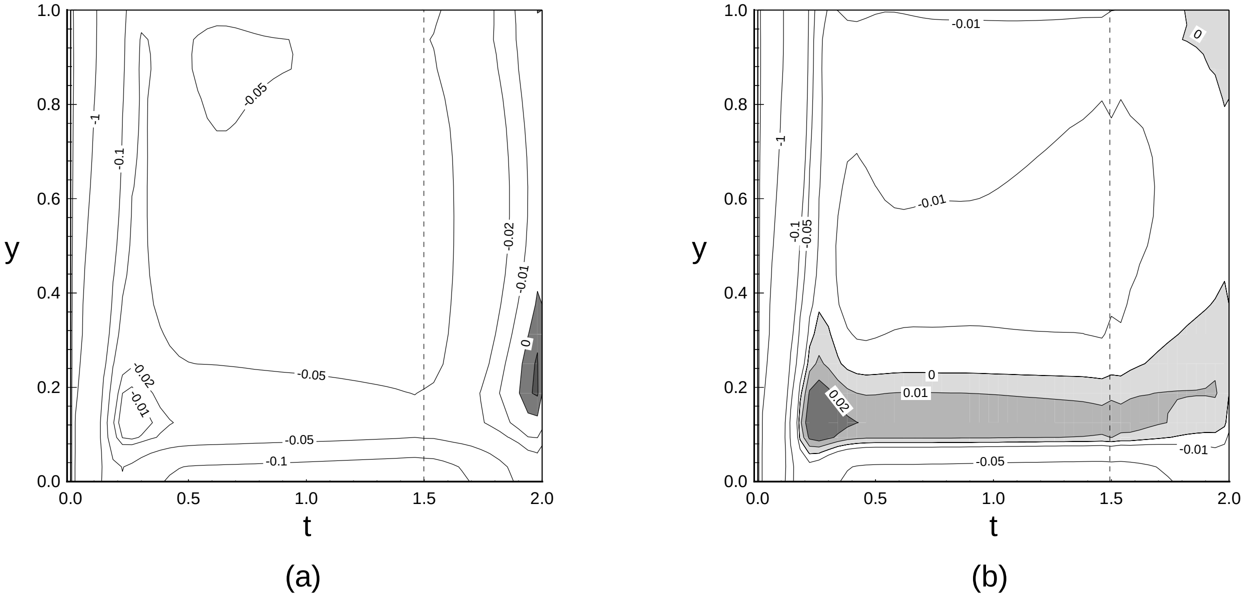

Figure 6: Contours of ΔG corresponding to the optimum $\bar{q}_{o\ell}(y,t)$: (a) $\delta = 0.2$: $\Delta G < 0$ which implies that a stable interface is achieved everywhere except in shaded region near $t = t_{max}$. The existence of the region is due to the fact that $\epsilon(t_{max}) = 0$ from $\boldsymbol{v}_f(t_{max}) = 0$ even though $\delta \neq 0$; (b) $\delta = 0$: a stripe (shaded) centering near the interface location $y = 0.12$ has $\Delta G > 0$ (unstable).

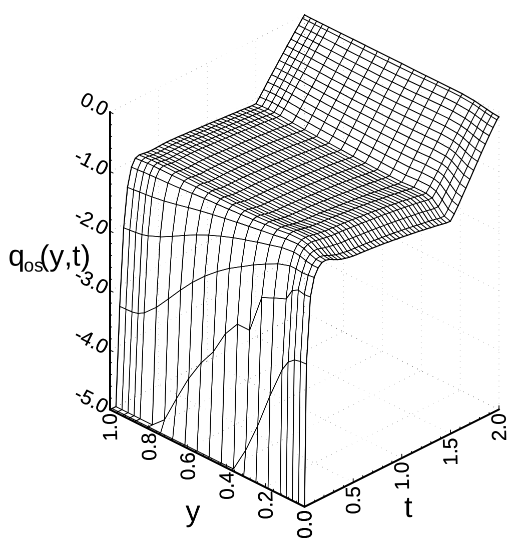

Figure 7: Optimum heat flux $q_{os}(y,t)$ at solid mold wall.