⊙ 研究論文

# Double – Diffusive Convection in Molten Pb – Sn Alloy

M. T. Hyun* · T. L. Bergman**

## 용융상태인 납-주석 합금의 이중확산유동

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**Key words** : Double – diffusive convection(이중확산유동), Chebyshev collocation technique (체비세프 콜로케이션 기법), Finite volume method(유한체적법), Buoyancy ratio (부력비)


## Abstract

액체 상태인 Pb – Sn 합금에 의한 이중확산유동에 대해 체비세프 콜로케이션 기법을 이용하여 수치해석하였다. 온도차에 의한 부력과 농도차에 의한 부력이 작을때에는 유동형태가 서서히 준정상상태에 이르러 아무런 진동현상을 볼 수 없다. 부력이 증가함에 따라 유동은 수직 농도 경계층을 파괴하여 플룸(Plume) 형태의 유동을 생성시키고, 이는 시스템 내부로 성장한 후 소멸된다. 이러한 현상이 반복되면서 높은 주파수의 진동현상을 관찰할 수 있다.


## Nomenclature

C : dimensionless concentration $=(C' - C_l)/(C_h - C_l)$

$C_h$ : concentration of high concentration wall, wt%

$C_l$ : concentration of low concentration wall, wt%

D : mass diffusivity, $m^2/s$

g : gravitational acceleration, $m/s^2$

$Gr_S$ : solutal Grashof number $=Ra \cdot N/Pr$

L : half length or height of the enclosure, m

N : buoyancy ratio $=\beta_C \cdot (C_h - C_l)/\beta_T \cdot (T_h - T_c)$

p : pressure, $N/m^2$

Pr : Prandtl number $=\nu/\alpha$

Ra : Rayleigh number $=g\beta_T(T_h - T_c)(2L)^3/(\nu \cdot \alpha)$

Sc : Schmidt number $=\nu/D$

Sh : local Sherwood number $=\partial C(t, -1, y)/\partial x$

Sh : mean Sherwood number $= \int_{-1}^{+1}(\partial C(t, -1, y)/\partial x)dy$

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T : dimensionless temperature $= (T' - T_c)/(T_h - T_c)$
$T_c$ : temperature of cold wall, ℃
$T_h$ : temperature of hot wall, ℃
t : dimensionless time $= t'\nu/L^2$
u : dimensionless x-direction velocity $= u'L/\nu$
v : dimensionless y-direction velocity $= v'L/\nu$
x : dimensionless horizontal coordinate $= x'/L$
y : dimensionless vertical coordinate $= y'/L$
$\alpha$ : thermal diffusivity, $m^2/s$
$\beta_C$ : coefficient of solutal expansion $= 1/\rho(\partial\rho/\partial C)_{T,p}$
$\beta_T$ : coefficient of thermal expansion $= -1/\rho(\partial\rho/\partial T)_{C,p}$
$\nu$ : kinematic viscosity, $m^2/s$
$\rho$ : density, $kg/m^3$
$\Psi$ : dimensionless stream function $= \Psi'/\nu$
$\omega$ : dimensionless vorticity $= \omega' L^2/\nu$

**Subscripts**

crit : critical
i : initial
max : maximum value
min : minimum value

**Superscript**

' : dimensional value

## 1. Introduction

Double-diffusive convection occurs in low Pr liquids during solidification of metals and semiconductor materials. It is well known that convection plays a major role in determining the species and crystalline distribution in the final product and, in turn, product properties. Simulation is motivated primarily by experimental difficulties involving low Pr liquids and, in the main, finite volume methods[1] have been used to predict the solidification process[2~5].

Although simulations have revealed interesting behavior, it is unclear whether the liquid phase transport is accurately predicted. Indeed, the extremely fine three dimensional structure and oscillatory behavior which has been observed experimentally in cavity convection involving moderate Pr, high Sc liquids[6,7] suggests that accurate simulation of thermosolutal convection at high $Gr_S$ is extremely difficult, if not impossible. In this study, liquid phase convection in molten Pb-Sn is simulated using a Chebyshev collocation(spectral) method. New double-diffusive phenomena is revealed at high $Gr_S$ with the direct simulation, and the predicted onset of oscillatory behavior is consistent with theory based upon experimental observation.

## 2. Mathematical Model

The system considered is two dimensional convection of Pb-Sn in a square enclosure of dimension 2L. The cavity top and bottom are insulated, while the left and right faces are held at uniform cold and hot temperature, respectively. All cavity walls are impermeable, but the left and right faces are maintained at Sn-rich(light) and Pb-rich(heavy) uniform concentrations. Hence, thermal(solutal) buoyacy forces induce counter clockwise(clockwise) circulation. Note that i) opposing thermosolutal liquid phase buoyancy forces are established during solidification of Pb-rich, Pb-Sn alloys and ii) fine three dimensional structure may be embedded within the overall two dimensional flow[6].

Preliminary studies showed that vorticity-velocity formulation of the convective behavior resulted in a faster numerical algorithm rela-

tive to stream function - vorticity or primitive variable formulations. Nearly identical predictions were generated with the different formulations, and the simulations reported here approximately 6 months CPU time on an IBM RS/6000 350 workstation. The governing equations in terms of vorticity - velocity functions are(with the origin at the cavity center) :

$$\partial^2 v/\partial x^2+\partial^2 v/\partial y^2=\partial\omega/\partial x \quad (1)$$

$$\partial^2 u/\partial x^2+\partial^2 u/\partial y^2=-\partial\omega/\partial y \quad (2)$$

$$8\cdot Pr\cdot((\partial\omega/\partial t+u\cdot\partial\omega/\partial x+v\cdot\partial\omega/\partial y)-(\partial^2\omega/\partial x^2+\partial^2\omega/\partial y^2))=Ra\cdot(\partial T/\partial x-N\cdot\partial C/\partial x) \quad (3)$$

$$Pr\cdot(\partial T/\partial t+u\cdot\partial T/\partial x+v\cdot\partial T/\partial y)=\partial^2 T/\partial x^2+\partial^2 T/\partial y^2 \quad (4)$$

$$Sc\cdot(\partial C/\partial t+u\cdot\partial C/\partial x+v\cdot\partial C/\partial y)=\partial^2 C/\partial x^2+\partial^2 C/\partial y^2 \quad (5)$$

The boundary and initial conditions are :

$$u(t,\pm1,y)=u(t,x,\pm1)=v(t,\pm1,y)=v(t,x,\pm1)=0 \quad (6a)$$

$$T(t,-1,y)=0,\ T(t,1,y)=1,\ \partial T(t,x,\pm1)/\partial y=0 \quad (6b)$$

$$C(t,-1,y)=1,\ C(t,1,y)=0,\ \partial C(t,x,\pm1)/\partial y=0 \quad (6c)$$

$$u(0,x,y)=v(0,x,y)=0,\ T(0,x,y)=0.5x+0.5,\ C(0,x,y)=0.5 \quad (6d)$$

Equations(1) - (5) were solved using a Chebyshev collocation technique[5]. In short, the dependent variable spatial distributions are expanded in doubly truncated series of Chebyshev polynomials using the usual Chebyshev - Gauss - Lobatto quadrature points[9]. Time intergration is achieved by a finite difference technique using the AB/second order backward Euler procedure, which combines implicit treatment of the diffusion terms(to improve numerical stability) with explicit treatment of the advection terms(to avoid iteration). This procedure, when applied to Eqs.(3) - (5), results in a two dimensional Helmholtz equation at each time level. A full matrix diagonalization algorithm[8] is used to solve the Helmholtz and Poisson Eqs.(1) and (2). Computational details are discussed in Madabhushi et al.[10] and Hyun et al.[11] and are not repeated here.

As shown by Hyun et al.[11], the predictions are insensitive to the time step($10^{-5}$ to $10^{-6}$) and spatial resolution($65\times65$ to $177\times177$ spatial modes) with a rapid and accelerating rate of convergence(of the $L^2$ - norm for the concentration field at a given time, as well as for the time - averaged $\overline{Sh}$ history) with spatial resolution. In addition, predictions have been compared to those of the finite volume method, and at Ra=5000, N= - 10 the two techniques produce highly disparate result[12]. The large discrepancy is attributed to numerical diffusion in the finite volume method which, in turn, dampens high frequency fluctuation.

## 3. Results

Simulations were performed for a lead - rich, Pb - Sn mixture. Although there is uncertainty regarding the thermophysical properties of this alloy[5], they were taken to be Pr=0.02 and Sc=150. Note that 8 and 18 equally spaced contours are drawn for the positive stream funtion and concentration distributions in Figs. 1 - 4, while negative stream funtion values are listed in the figure captions. Since they are not interesting, temperature distributions are not shown.

Selected concentration and stream funtion distributions for Ra=100, N= - 10 are shown in Fig. 1. Under conditions of low thermal buoyancy, the system response consists of Pb - (Sn - ) rich liquid falling(rising) along the right(left) cavity face with solutally - stratified conditions eventually existing throughout the enclosure

(Fig. 1d). Thermal buoyancy forces induce counter clockwise core rotation and, with time the core shrinks due to interaction with the clockwise rotation associated with solutal buoyancy forces.

Relative to the response at low buoyancy, conditions for Ra=5000, N= - 10(Fig. 2) show fundamentally different behavior. The concentration distributions are complicated by a thermally - affected entrainment process along the edge of the species boundary layers at, approxi-

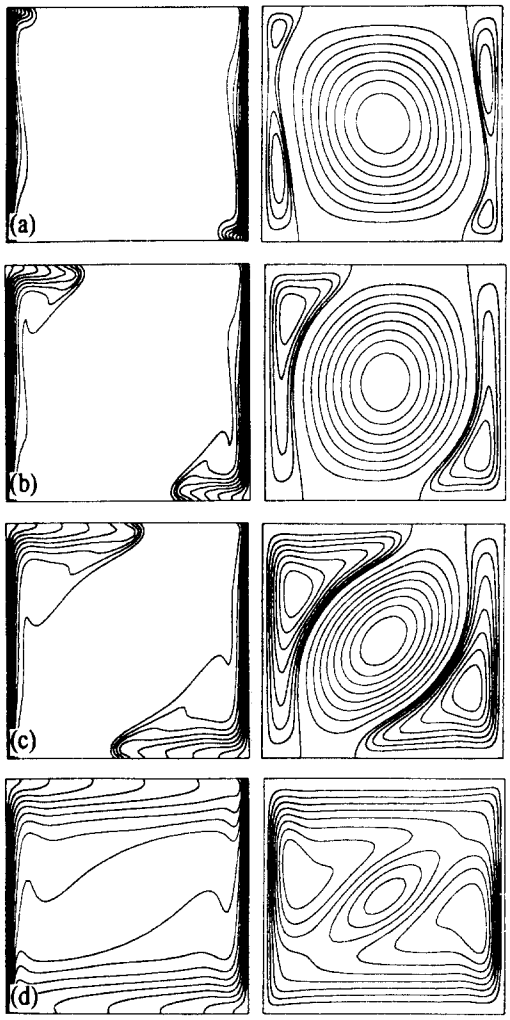


**Fig. 1 Predicted concentration and stream function distibutions for Ra=100 and N= - 10 at (a) t=0.3($\psi_{max}$=4.94, $\psi_{min}$= - 0.189, $\psi$= - 0.047, - 0.095, - 0.142),(b) t=0.75($\psi_{max}$= 3.895, $\psi_{min}$= - 0.385, $\psi$= - 0.077, - 0.154, - 0.231, - 0.308),(c) t=1.35($\psi_{max}$=1.883, $\psi_{min}$= - 0.466, $\psi$= - 0.058, - 0.117, 0.175, - 0.233, - 0.291, - 0.350, - 0.408),(d) t= 3.15($\psi_{max}$=0.321, $\psi_{min}$=0).**

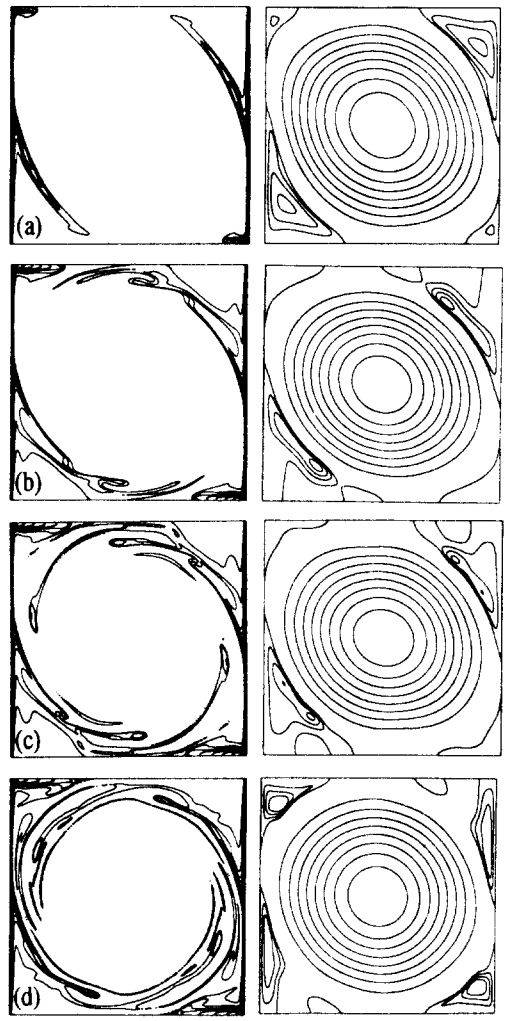


**Fig. 2 Predicted concentration and stream function distributions for Ra=5000 and N= - 10 at (a) t=0.06($\psi_{max}$=124.3, $\psi_{min}$= - 0.97, $\psi$= 0.3, 0.6, 0.9),(b) t=0.14($\psi_{max}$=141.7, $\psi_{min}$= - 2.1, $\psi$= - 0.6, - 1.2, - 1.8),(c) t= 0.17($\psi_{max}$=144.7, $\psi_{min}$= - 1.6, $\psi$= 0.5, 1.0, 1.5),(d) t=0.22($\psi_{max}$=152.2, $\psi_{min}$= 0.89, $\psi$= - 0.2, - 0.4, - 0.6).**

mately, cavity mid - height(Fig. 2a). Rather than being smoothly incorporated into the core, the entrained liquid is broken into small drops which are subsequently advected thermally throughout the domain. Breakup times are random and, as will be shown, the intermittent nature of the flow leads to a high frequency temporal variation in the $\overline{Sh}$ history. Droplet trajectory dominated by its solutal buoyancy force component, as observed experimentally by Jiang et al.[6], was not noted here. A much less robust, yet intermittent entrainment phenomena was found at Ra= 1000, N= - 10(Ref. 11). Simulation of the Ra= 5000, N= - 10 case using the finite volume method produced no high frequency behavior or solutal boundary layer breakup by Hyun et al.[12].

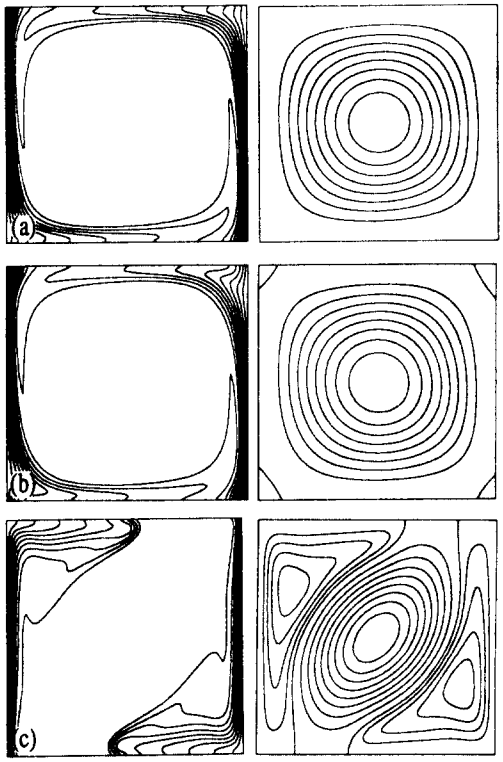


**Fig. 3 Predicted concentration and stream function distributions for Ra=100 and t=1.35 : (a) N= - 0.1($\psi_{max}$=6.305, $\psi_{min}$=0), (b) N= - 1($\psi_{max}$=6.20, $\psi_{min}$= - 0.003), (c) N= - 10 ($\psi_{max}$=1.883, $\psi_{min}$= - 0.466, $\psi$= - 0.1, - 0.2, - 0.3, - 0.4).**

Figures 3 and 4 include predicted behavior at particular times for Ra=100(Fig. 3) and 5000 (Fig. 4) under a range of solutal buoyancy conditions. At either Ra value, increasing N from - 0.1 to - 1 modestly affects the system response. Only slight variation in the concentration distribution is noted, along with the establishment of corner recirculations at N= - 1. As N is increased further to - 10(Figs. 3c and 4c), however, transition to overall downflow(upflow) along the right(left) wall is noted along with advancing solutal fronts at the cavity ceiling and floor in Fig. 3c, while a complex species distribution is evident in Fig. 4c.

Local Sh distributions are reported in Fig. 5

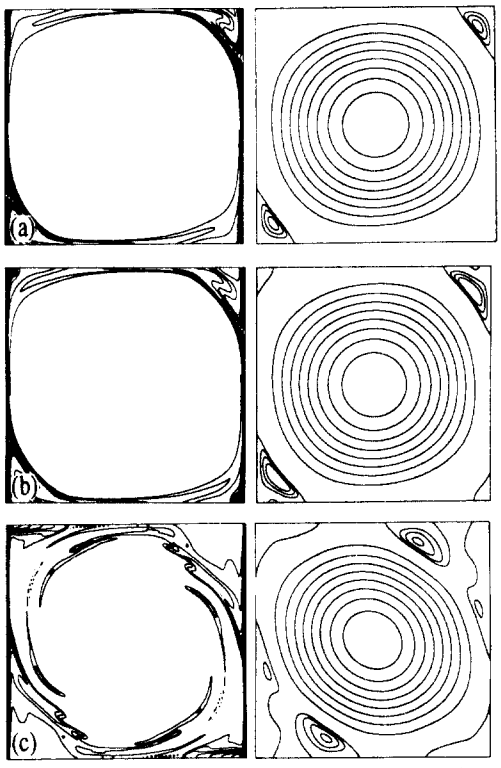


**Fig. 4 Predicted concentration and stream function distributions for Ra=5000 and t=0.18 : (a) N= - 0.1($\psi_{max}$=170.0, $\psi_{min}$= - 0.249, $\psi$= - 0.07, - 0.14, - 0.21),(b) N= - 1($\psi_{max}$=170.0, $\psi_{min}$= - 0.440, $\psi$= - 0.1, - 0.2, - 0.3), (c) N= - 10($\psi_{max}$=146.2, $\psi_{min}$= - 3.260, $\psi$= - 1.0, - 2.0, - 3.0).**

for the Ra=100 and 5000 results of Figs. 3 and 4(Figs. 5a and 5b, respectively). At low Ra, significant solutal buoyancy forces(N= - 10) trigger a severe redistribution in local species transport rates in response to the wall flow reversal noted in Fig. 3c. Local Sh minima are evident in Fig. 5b, and coincide with wall flow stagnation regions most notably evident in Fig. 4c. Note that i) wall stagnation regions have been observed exeperimentally[6,7] and ii) the $\overline{Sh}$ distribution is not smooth at Ra=5000, N= - 10 because of the intermittent breakup of the solutal boundary layer.

Because of the tenacity with which liquids retain their solutal identity(high Sc), interpretation of the boundary layer breakup and entrainment process is difficult if $0 \leq C \leq 1$ is plotted. Figure 6 includes a series of concentration contours for $0 \leq C \leq 0.45$(Pb - rich liquid).

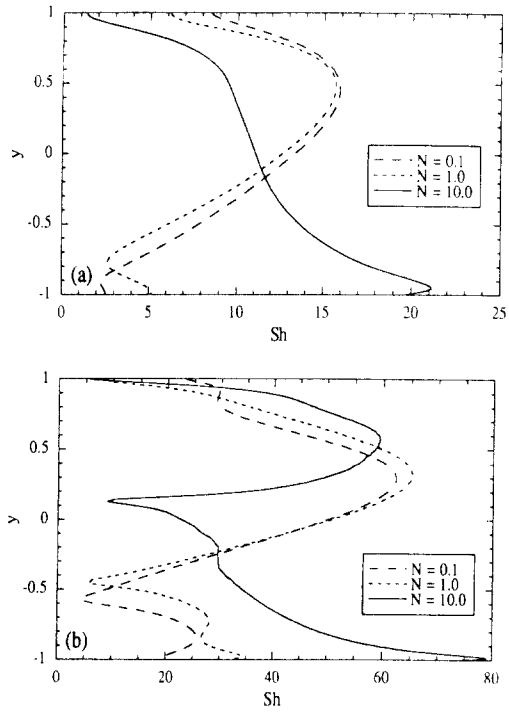


**Fig. 5 Local Sh with buoyancy ratio along left wall : (a) Ra=100 and t=1.35, (b) Ra=5000 and t=0.18.**

Close inspection shows that i) droplet formation dynamics are extremely complex, and are different for separate droplets(compare drops 2 and 4), ii) droplet formation is affected by proximity to the cavity ceiling(drop 3), iii) the droplet life span varies(compare drops 1 and 2), iv) the droplet shape is affected by vorticity (drop 1, Figs. 6f, g and h) and v) the frequency of drop formation is very high compared to the rate at which the Pb - rich fluid falls along the right wall to the cavity floor. The influence of the drop longevity(drops of Pb - rich liquid are injected into cold fluid regions, resulting in significant local subcooling relative to the material's liquidus) on potential solidification dynamics is quantified in Hyun et al.[12].

Selected Sh histories are shown in Fig. 7. Since $C_i=0.5$, $\overline{Sh}$ is initially infinite and predictions are insensitive to buoyancy forces until convective motion is established. In each case, predicted histories are similar for N= - 0.1 and - 1, with major variations associated with N= - 10. At Ra=1000(Fig. 7b) Sh is reduced at N= - 10 due to i) the establishment of a solutally - inactive core region and ii) absence of significant high frequency mixing as shown in Hyun et al.[11]. In contrast, Sh is enhanced at N= - 10 for Ra=100(Fig. 7a) and Ra=5000 (Fig. 7c) by i) overall flow reversal and the "filling box" behavior at small Ra and ii) establishment of high frequency mixing at large Ra.

To further examine the high frequency behavior, Fig. 7d shows part of the Sh histories for i) Ra=5000, N= - 10 and ii) Ra=2500, N= - 20. These extreme, $Gr_S^{1/4} \approx 40$ predictions are in the experimental range of Jiang et al.[7], and the values of N bracket $N_{crit}$ which marks the transition between their fluctuating and quasi - steady experimental results. Consistent with the experimentation, the Ra=5000 case is characterized by high frequency fluctuation,

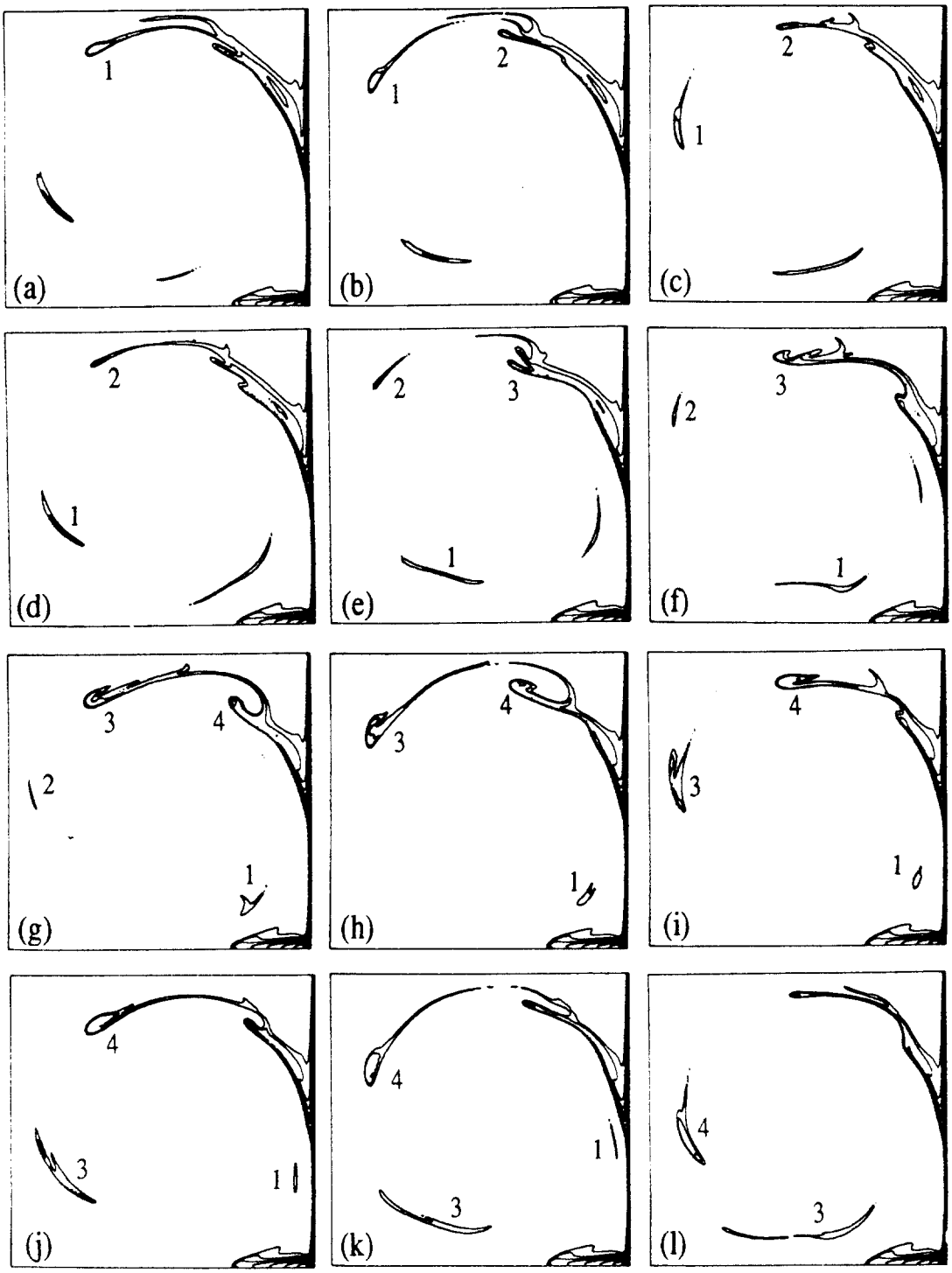


**Fig. 6 Time series of Pb rich concentration contours. The initial is t=0.175 and results are shown at time increments of 0.0025. Contour values are shown for $0 \leq C \leq 0.45$ in increments of 0.05. Individual drops are identified.**

while the Ra=2500 case experiences only modest, low frequency variation with time. Considering that i) the experiments were performed with a moderate Pr liquid in shallow enclosures, and ii) the observed flow was characterized by fine three dimensional structure, it is remarkable that present two dimensional simulations exhibit transition to high frequency, thermosolutally - driven oscillatory behavior in a manner with the experimentally - based theory.

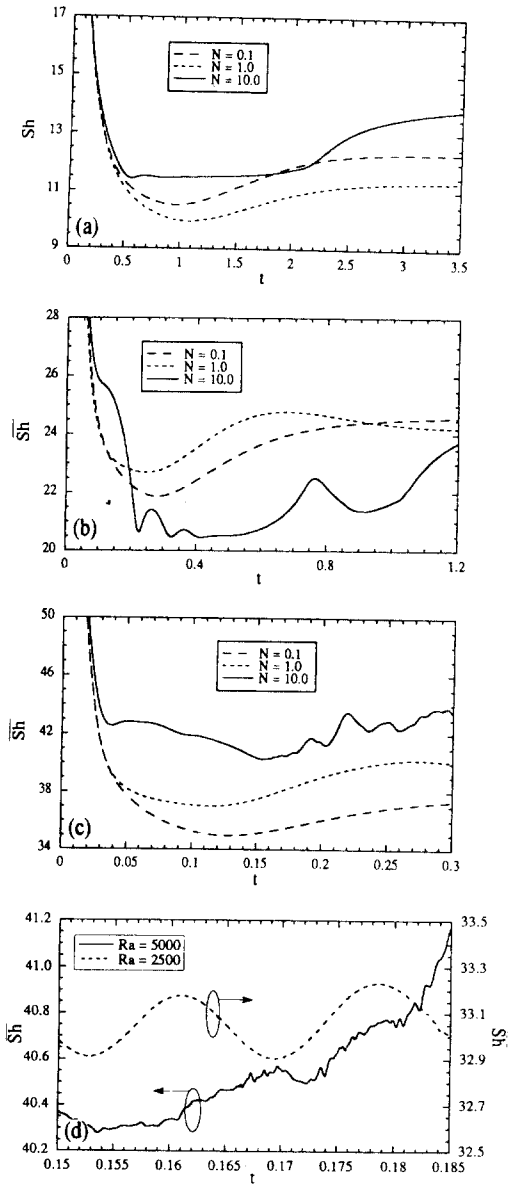


**Fig. 7 Predicted Sh histories with buoyancy ratio : (a) Ra=100, (b) Ra=1000, (c) Ra=5000, (d) detailed histories for Ra=5000 and 2500.**

## 4. Summary and Conclusions

In this study, a spectral method was used to predict two dimensional double - diffusive convection in a low Pr liquid. Under some conditions, a highly complex yet realistic concentration distribution is induced by thermal convection which triggers high frequency system response related to the breakdown of the solutal boundary layer. The transition from quasi - steady to high frequency oscillatory convection is consistent with the criterion developed by Jinang et al.[7]. Since finite volume method simulations produced no transition, and since most solidification simulations employ the finite volume method, it is probable that some solidification simulations do not adequately predict relevant liquid phase convection.

Features which might be addressed include the effects of three - dimensionality and response in other aspect ratio enclosures. Solidification dynamics may be included, although the details concerning their inclusion in a direct simulation are unclear at this time. Experimental validation of the predicted complex concentration distributions in a low Pr melt is desirable.

## Acknowledgement

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