

Double-diffusive convection affected by conductive and insulating side walls during physical vapor transport of Hg_2Br_2

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Abstract In last few decades, although thermal and/or solutal buoyancy-driven recirculating flows in a closed ampoule have been intensively studied as a model problem, there exist interesting total molar flux of Hg_2Br_2 that have been unreported in the literature. It is concluded that the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ increases linearly and directly as the temperature difference regions in the range of $10^\circ\text{C} \leq \Delta T \leq 50^\circ$, $3.5 \times 10^3 \leq \text{Gr}_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq \text{Gr}_s \leq 6.87 \times 10^4$. For the range of $10 \text{ Torr} \leq P_B \leq 150 \text{ Torr}$, the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ decays second order exponentially as the partial pressure of component B (argon as an impurity), P_B increases. From the view point of energy transport, the fewer the partial pressure of component B (argon), P_B is, the more the energy transport is achieved.

Key words Double diffusion, Physical vapor transport, Hg_2Br_2

1. Introduction

During the past few decades one area of interest in double diffusion has been the study of the physical vapor transport (PVT) processes in a sealed chamber. In recent years, the problems of Cattaneo-Christov double diffusion have been studied for Williamson nanomaterials slip flow subject to porous medium [1], and bi-directional stretched nanofluid flow with Cattaneo-Christov double diffusion [2]. Muhammad et al. [3] addressed Darcy-Forchheimer flow over an exponentially stretching curved surface with Cattaneo-Christov double diffusion. Asha and Sunitha [4] reported thermal radiation and hall effects on peristaltic blood flow with double diffusion in the presence of nanoparticles.

Mercurous halide materials are well known as the most promising materials in applications for acousto-optic materials and signal processing optics, for example, Bragg cells. Singh and his group investigated systematically the growth and characterization and development of large single crystals of Hg_2Br_2 [5-11]. Many reports of Hg_2Br_2 could be found in references [12-16]. Kim and his coworkers [17-19] have performed two-dimensional numerical studies of double diffusion convection in the vapor phase during physical vapor crystal growth. Duval [20] published that four flow structure

regions appear during the physical vapor transport of mercurous chloride crystal growth.

Our numerical simulations are motivated by the desire to study the influences of hybrid thermal boundary conditions on the convective flow because the final quality of crystal is affected by convection fields. In this paper, the effects of the temperature differences between the source and crystal, the Peclet number, Pe , and the partial pressure of component B (argon as inert gas), P_B on the total molar flux of Hg_2Br_2 and the maximum magnitudes of velocity vector, $|U|_{\text{max}}$ in the dimensional unit (cm/sec) shall be addressed.

2. Numerical Simulations

Consider steady state thermal and solutal buoyancy driven recirculating flows of $\text{Hg}_2\text{Br}_2(\text{A})$ -argon (B) with thermo-physical properties listed in Table 1, in PVT crystal growth enclosure for hybrid thermal boundary conditions with linear temperature profiles, i.e., conductive walls, and insulating walls, shown in Fig. 1, accompanied by a 42×22 ($x \times y$) grid system. The detailed assumptions and nomenclature can be found in reference [17]. Also, the dimensionless parameters of Prandtl, Lewis, Peclet, Grashof, concentration, aspect ratio are described in reference [20].

In non-dimensional form, continuity, Navier-Stokes momenta, energy transport, and mass transport are gov-

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Table 1
Thermo-physical properties of Hg₂Br₂(A)-argon (B) (M_A = 560.988, M_B = 39.944) at ΔT = 50°C, P_B = 10 Torr

Kinematic viscosity	0.44 cm ² /sec
Thermal diffusivity	0.45 cm ² /sec ²
Binary diffusivity	1.28 cm ² /sec ²
Coefficient of thermal volume expansion	0.0017 (1/°C)
Density of mixture	0.000599 g/cm ³
Prandtl number	0.97
Lewis number	0.35
Peclet number	3.65
Concentration number	1.02
Thermal Grashof number	3.5 × 10 ³
Solutal Grashof number	4.94 × 10 ⁴

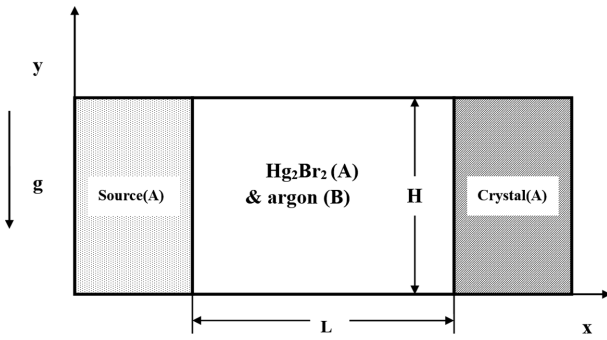


Fig. 1. System schematic and coordinates for numerical simulation of PVT crystal growth reactor of Hg₂Br₂(A)-argon (B).

governed by:

$$\nabla^* \bullet \mathbf{V} = 0, \quad (1)$$

$$\vec{\nabla} \bullet \nabla^* \vec{\mathbf{V}} = -\nabla^* p^* + \text{Pr} \nabla^{*2} \vec{\mathbf{V}} - \frac{\text{Gr} \cdot \text{Pr}^2}{\text{Ar}^3} \frac{(1 - \rho^*)}{\beta \Delta T}, \quad (2)$$

$$\vec{\nabla} \bullet \nabla^* T^* = \nabla^{*2} T^*, \quad (3)$$

$$\vec{\nabla} \bullet \nabla^* \omega_A^* = \frac{1}{\text{Le}} \nabla^{*2} \omega_A^*. \quad (4)$$

The boundary conditions corresponding to the above equations (1) to (4) are given as follows:

On the walls

$$(0 < x^* < 1, y^* = 0 \text{ and } 1):$$

$$u(x^*, 0) = u(x^*, 1) = v(x^*, 0) = v(x^*, 1) = 0 \quad (5)$$

$$\frac{\partial \omega_A^*(x^*, 0)}{\partial y^*} = \frac{\partial \omega_A^*(x^*, 1)}{\partial y^*} = 0,$$

$$T^*(x^*, 0) = -\frac{1}{\text{Ar}} \cdot x^* + 1$$

$$(1 < x^* < 3, y^* = 0 \text{ and } 1):$$

$$u(x^*, 0) = u(x^*, 1) = v(x^*, 0) = v(x^*, 1) = 0 \quad (6)$$

$$\frac{\partial \omega_A^*(x^*, 0)}{\partial y^*} = \frac{\partial \omega_A^*(x^*, 1)}{\partial y^*} = 0,$$

$$\frac{\partial T^*(x^*, 0)}{\partial y^*} = \frac{\partial T^*(x^*, 1)}{\partial y^*} = 0$$

$$(3 < x^* < 4, y^* = 0 \text{ and } 1):$$

$$u(x^*, 0) = u(x^*, 1) = v(x^*, 0) = v(x^*, 1) = 0 \quad (7)$$

$$\frac{\partial \omega_A^*(x^*, 0)}{\partial y^*} = \frac{\partial \omega_A^*(x^*, 1)}{\partial y^*} = 0,$$

$$T^*(x^*, 0) = -\frac{1}{\text{Ar}} \cdot x^* + 1$$

On the source ($x^* = 0, 0 < y^* < 1$):

$$u(0, y^*) = -\frac{1}{\text{Le}} \frac{1}{(\text{Cv} - 1)} \frac{\partial \omega_A^*(0, y^*)}{\partial x^*}, \quad (8)$$

$$v(0, y^*) = 0,$$

$$T^*(0, y^*) = 1,$$

$$\omega_A^*(0, y^*) = 1.$$

On the crystal ($x^* = L/H, 0 < y^* < 1$):

$$u(L/H, y^*) = -\frac{1}{\text{Le}} \frac{1}{\text{Cv}} \frac{\partial \omega_A^*(L/H, y^*)}{\partial x^*}, \quad (9)$$

$$v(L/H, y^*) = 0,$$

$$T^*(L/H, y^*) = 0,$$

$$\omega_A^*(L/H, y^*) = 0.$$

The code verification of one's results can be found in reference [17], for the Semi-Implicit Method Pressure-Linked Equations Revised (SIMPLER) [21].

3. Results and Discussion

When $M_A \neq M_B$, the two molecular weights of Hg₂Br₂ and argon are different, i.e., $M_A = 560.988$ g/gmol, $M_B = 39.944$ g/gmol, solutally buoyancy driven convection is much important compared with thermally buoyancy driven convection, but solutal and/or thermal convection are coupled and the effects of thermal buoyancy convection cannot be neglected during the physical vapor transport of Hg₂Br₂ in the vapor phase. Therefore, our interest is restricted on our studies to investigate the relations of the driving force, the temperature difference, ΔT, the maximum magnitudes of velocity vector, $|U|_{\text{max}}$ and the Peclet number for the transport of crystal species.

As shown in Fig. 2, it is clear that the total molar flux of Hg₂Br₂(A) increases linearly and directly as the temperature difference in the range of $10^\circ\text{C} \leq \Delta T \leq 50^\circ\text{C}$,

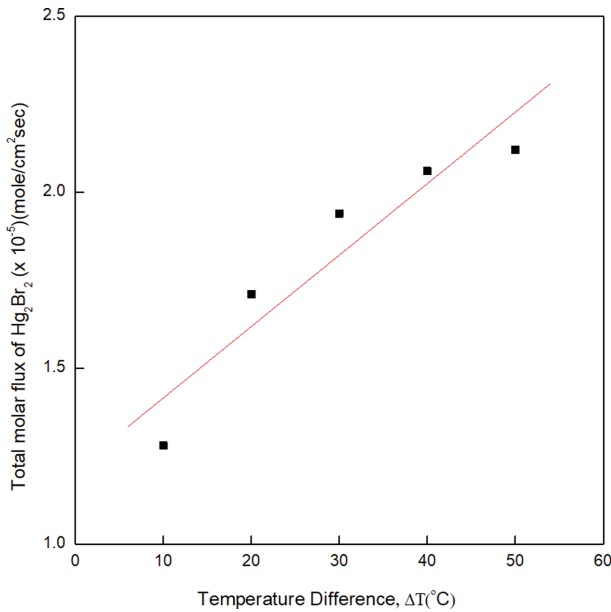


Fig. 2. The total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ as a function of the temperature difference, ΔT ($^{\circ}\text{C}$), based on aspect ratio = 4, $T_s = 300^{\circ}\text{C}$, $P_B = 10$ Torr, $3.5 \times 10^3 \leq Gr_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq Gr_s \leq 6.87 \times 10^4$, on earth.

$3.5 \times 10^3 \leq Gr_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq Gr_s \leq 6.87 \times 10^4$. For $\Delta T = 10^{\circ}\text{C}$, the corresponding thermal (Gr_t) and solutal (Gr_s) Grashof number is 4.08×10^3 and 6.87×10^4 , respectively; for $\Delta T = 50^{\circ}\text{C}$, the corresponding thermal (Gr_t) and solutal (Gr_s) Grashof number is 3.5×10^3 and 4.94×10^4 , respectively. With increasing the temperature difference, the corresponding thermal (Gr_t) and solutal (Gr_s) Grashof number reversely decrease, which reflected the variations in the density of the mixture of Hg_2Br_2 and argon. In other words, for $\Delta T = 10^{\circ}\text{C}$, the kinematic viscosity is $0.18 \text{ cm}^2/\text{sec}$; for $\Delta T = 50^{\circ}\text{C}$, the kinematic viscosity is $0.44 \text{ cm}^2/\text{sec}$. The system considered in Fig. 1 is Ar (aspect ratio, transport length-to-width) = 4, T_s (source temperature) = 300°C , P_B (partial pressure of component B, argon) = 10 Torr, on earth. For $\Delta T = 10^{\circ}\text{C}$, the thermal diffusivity and binary mass diffusivity is 0.18, and $1.28 \text{ cm}^2/\text{sec}$; for $\Delta T = 50^{\circ}\text{C}$, the thermal diffusivity and binary mass diffusivity is 0.45, and $1.28 \text{ cm}^2/\text{sec}$. For the range of $10^{\circ}\text{C} \leq \Delta T \leq 30^{\circ}\text{C}$, the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ increases sharply with increasing the temperature difference, whereas for the range of $30^{\circ}\text{C} \leq \Delta T \leq 50^{\circ}\text{C}$, the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ increases relatively gradually. As mentioned before, this difference is likely be due to the variations in the density of the mixture of Hg_2Br_2 and argon.

Figure 3 illustrates the effects of Peclet number, Pe on as the temperature difference, ΔT in the range of $10^{\circ}\text{C} \leq \Delta T \leq 50^{\circ}\text{C}$, $3.5 \times 10^3 \leq Gr_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq Gr_s \leq$

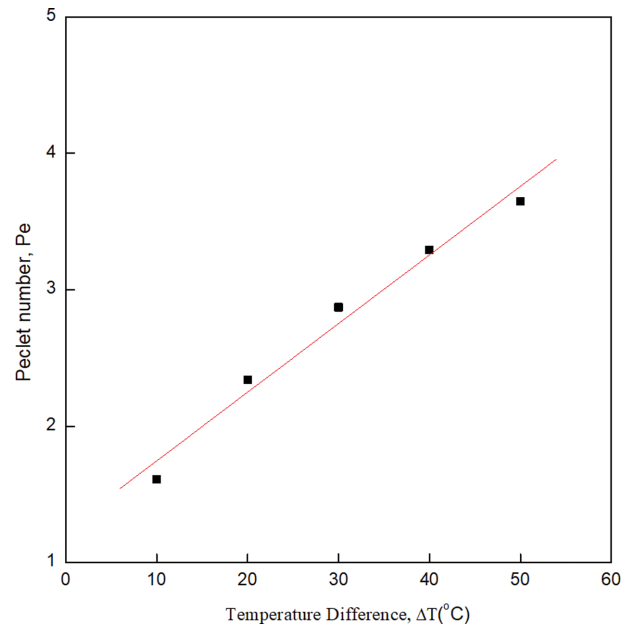


Fig. 3. The Peclet number, Pe as a function of the temperature difference, ΔT ($^{\circ}\text{C}$), based on aspect ratio = 4, $T_s = 300^{\circ}\text{C}$, $P_B = 10$ Torr, $3.5 \times 10^3 \leq Gr_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq Gr_s \leq 6.87 \times 10^4$, on earth.

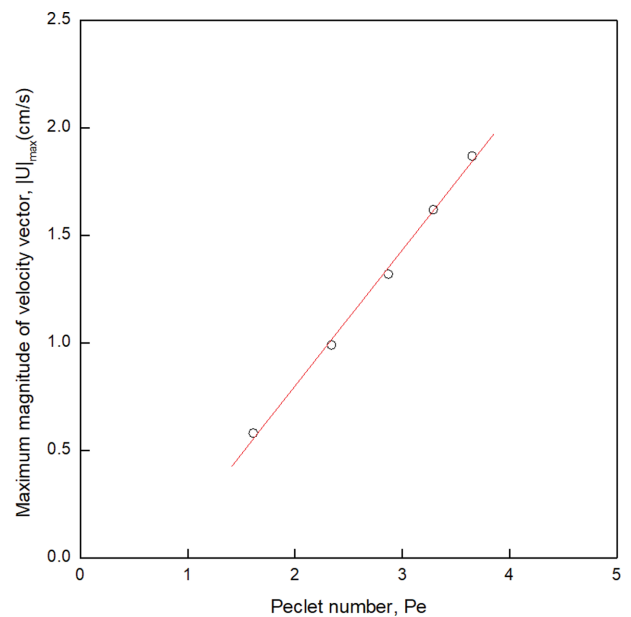


Fig. 4. The $|U|_{\max}$ as a function of the dimensionless Peclet number, Pe , based on aspect ratio = 4, $T_s = 300^{\circ}\text{C}$, $P_B = 10$ Torr, $3.5 \times 10^3 \leq Gr_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq Gr_s \leq 6.87 \times 10^4$, on earth.

6.87×10^4 . The Peclet number, Pe increases linearly and directly with the temperature differences. Figure 4 shows the relation of the maximum magnitude of velocity vector, $|U|_{\max}$ and Peclet number, Pe for $10^{\circ}\text{C} \leq \Delta T \leq 50^{\circ}\text{C}$, aspect ratio = 4, $T_s = 300^{\circ}\text{C}$, $P_B = 10$ Torr, on earth. This relation illustrates that the $|U|_{\max}$ increases linearly with the Peclet number, Pe . Note that the Peclet number is

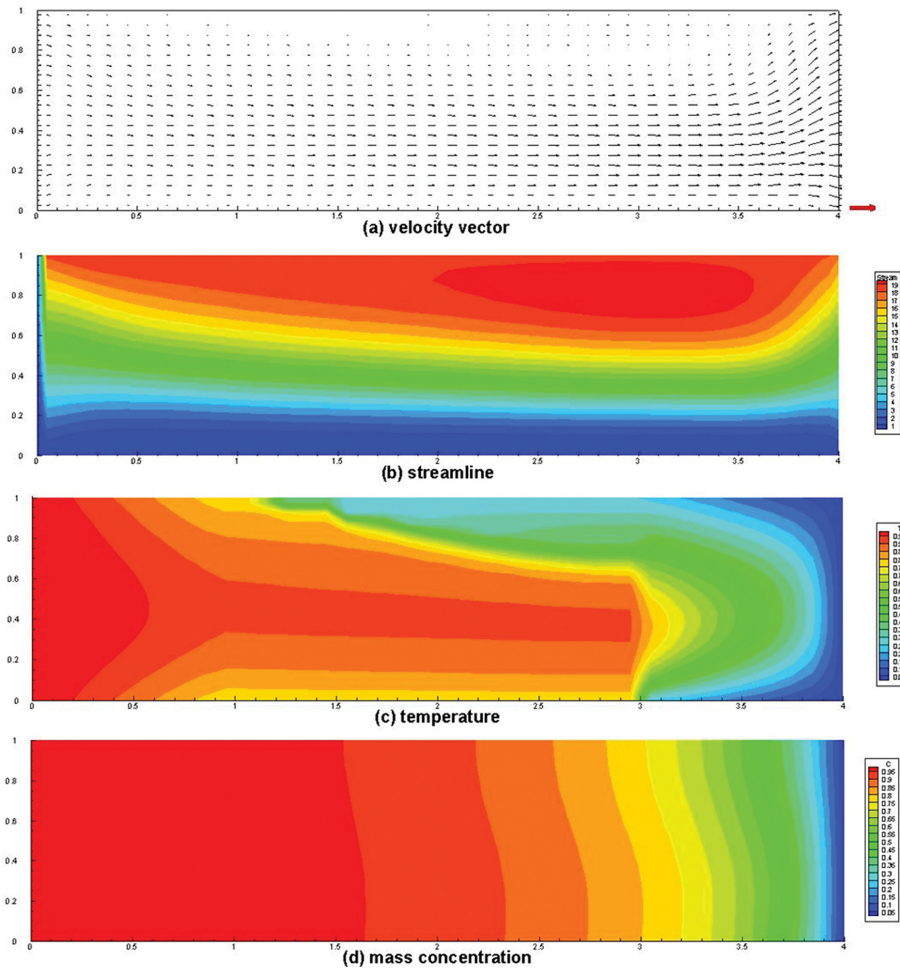


Fig. 5. (a) Velocity vector, (b) streamline, (c) temperature, (d) mass concentration profile, based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$ ($300^\circ\text{C} \rightarrow 250^\circ\text{C}$), $P_B = 10$ Torr, Peclet number = 3.65, thermal Grashof number (Gr_t) = 3.5×10^3 , solutal Grashof number (Gr_c) = 4.94×10^4 , Prandtl number = 0.97, Lewis number = 0.35, concentration parameter = 1.02, total pressure = 108 Torr, $|U|_{\max} = 1.87$ cm/sec, on earth.

intimately related to the advectations across the interfaces at the source and the crystal regions.

Figure 5 shows the profiles of velocity vector, streamline, temperature, mass concentration, based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$ ($300^\circ\text{C} \rightarrow 250^\circ\text{C}$), $P_B = 10$ Torr, $|U|_{\max} = 1.87$ cm/sec, on earth. As plotted in Fig. 5, there exists small one convective cell in the vapor phase, and the flow structure is asymmetrical against at $y^* = 0.5$ and three-dimensional flow structure. For the flow regions along the transport length at the bottom region, i.e., $0 \leq y^* \leq 0.5$, the one-dimensional Stefan flows appear. Temperature profile shown in Fig. 5(c) is related to the hybrid thermal boundary conditions; for $0 \leq x^* \leq 1$, conductive walls and for $1 \leq x^* \leq 3$, insulating walls, for $3 \leq x^* \leq 4$, conductive walls. Close spacings of mass concentration shown in Fig. 5(d) exhibits the mechanism of the diffusion-limited mass transfer.

Figure 6 shows the effects of the partial pressure of

component B (argon), P_B , on the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$, for $10 \text{ Torr} \leq P_B \leq 150 \text{ Torr}$, based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$, $T_s = 300^\circ\text{C}$, $P_B = 10$ Torr, $1.8 \times 10^3 \leq Gr_t \leq 2.9 \times 10^3$, $2.6 \times 10^4 \leq Gr_s \leq 4.9 \times 10^4$, on earth. For the range of $10 \text{ Torr} \leq P_B \leq 150 \text{ Torr}$, the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ decays second order exponentially with the partial pressure of component B (argon), P_B . Figure 7 shows the influences of the partial pressure of component B (argon), P_B , on the Peclet number, Pe , for $10 \text{ Torr} \leq P_B \leq 150 \text{ Torr}$, corresponding to Fig. 6.

Figure 8 shows the profiles of velocity vector, streamline, temperature, mass concentration, based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$ ($300^\circ\text{C} \rightarrow 250^\circ\text{C}$), $P_B = 60$ Torr, $|U|_{\max} = 0.99$ cm/sec, on earth. As shown in Fig. 8, one convective roll is present in the vapor phase. In a comparison of Fig. 8(c) temperature with Fig. 5(c) temperature, from the view point of energy transport, the fewer the partial pressure of component B (argon), P_B is, the more

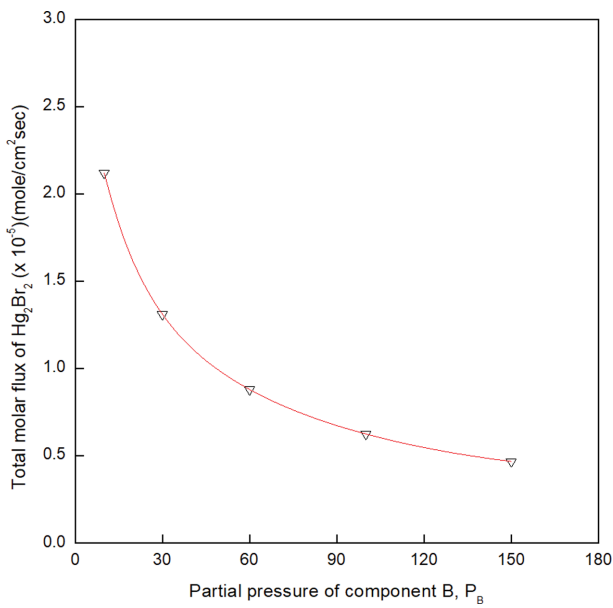


Fig. 6. The total molar flux of Hg_2Br_2 (A) as the partial pressure of component B, P_B , based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$, $T_s = 300^\circ\text{C}$, $P_B = 10$ Torr, $1.8 \times 10^3 \leq Gr_t \leq 2.9 \times 10^3$, $2.6 \times 10^4 \leq Gr_s \leq 4.9 \times 10^4$, on earth.

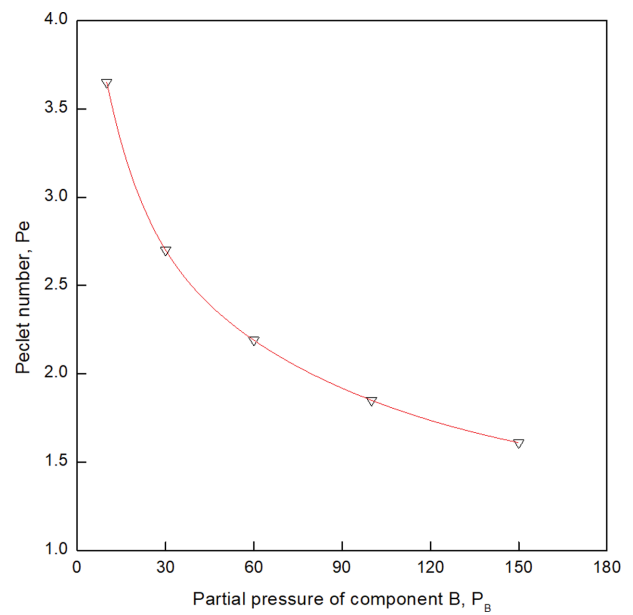


Fig. 7. The Peclet number, Pe as the partial pressure of component B, P_B , ΔT ($^\circ\text{C}$), based on aspect ratio = 4, $T_s = 300^\circ\text{C}$, $P_B = 10$ Torr, $3.5 \times 10^3 \leq Gr_t \leq 4.6 \times 10^3$, $4.9 \times 10^4 \leq Gr_s \leq 5.6 \times 10^4$, on earth.

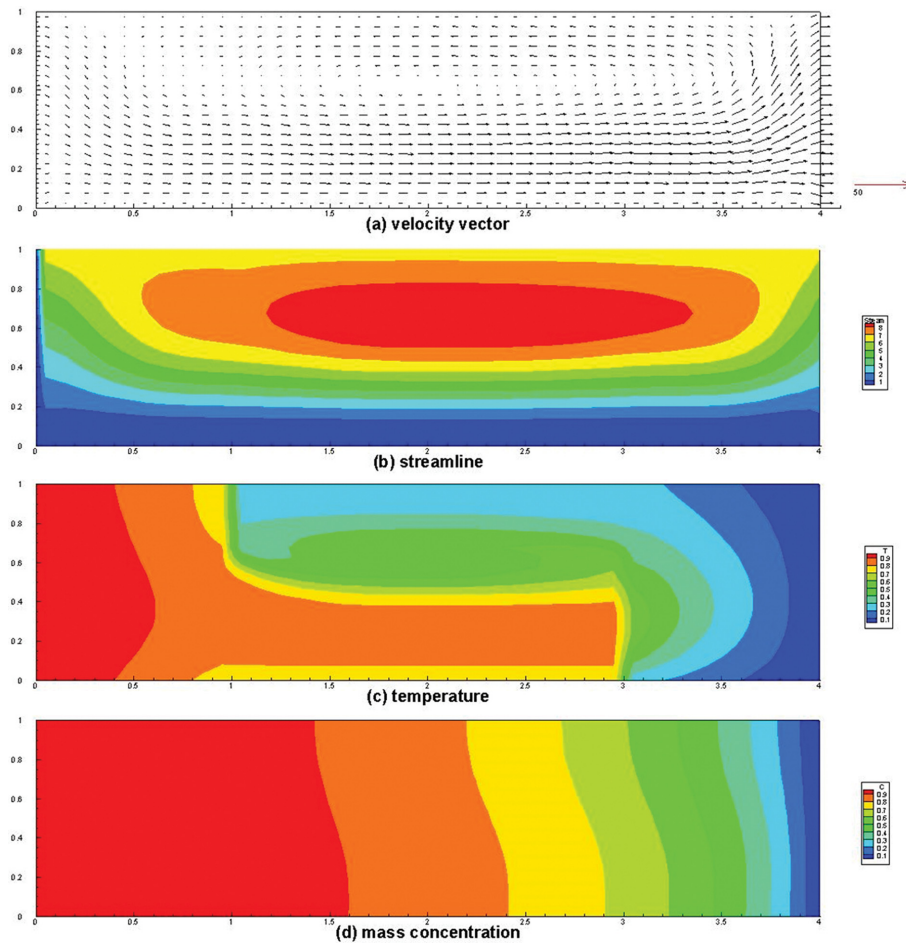


Fig. 8. (a) Velocity vector, (b) streamline, (c) temperature, (d) mass concentration profile, based on aspect ratio = 4, $\Delta T = 50^\circ\text{C}$ ($300^\circ\text{C} \rightarrow 250^\circ\text{C}$), $P_B = 60$ Torr, Peclet number = 2.19, thermal Grashof number (Gr_t) = 3.8×10^3 , solutal Grashof number (Gr_s) = 5.07×10^4 , Prandtl number = 0.92, Lewis number = 0.51, concentration parameter = 1.12, total pressure = 158 Torr, $|U|_{\max} = 0.99$ cm/sec, on earth.

the energy transport is achieved.

4. Conclusions

It is concluded that the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ increases linearly and directly as the temperature difference, ΔT in the range of $10^\circ\text{C} \leq \Delta T \leq 50^\circ\text{C}$, $3.5 \times 10^3 \leq \text{Gr}_t \leq 4.08 \times 10^3$, $4.94 \times 10^4 \leq \text{Gr}_s \leq 6.87 \times 10^4$. For the range of $10 \text{ Torr} \leq P_b \leq 150 \text{ Torr}$, the total molar flux of $\text{Hg}_2\text{Br}_2(\text{A})$ decays second order exponentially with the partial pressure of component B (argon), P_b . From the view point of energy transport, the fewer the partial pressure of component B (argon), P_b is, the more the energy transport is achieved.

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