Lattice Boltzmann Modeling of the Dissolution Process of Silicon into Germanium using a Simplified Crystal Growth Technique

Farid Mechighel, Niel Armour, Sadik Dost, Mohammed El Ganaoui, Mahfoud Kadja

December 2017

Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 139 (2017) 147–152

Energy Procedia

www.elsevier.com/locate/procedia

International Conference On Materials And Energy 2015, ICOME 15, 19-22 May 2015, Tetouan, Morocco, and the International Conference On Materials And Energy 2016, ICOME 16, 17-20 May 2016, La Rochelle, France

Lattice Boltzmann Modeling of the Dissolution Process of Silicon into Germanium using a Simplified Crystal Growth Technique

Farid Mechighel[a,b,*] Niel Armour[b], Sadik Dost[b], Mohammed El Ganaoui[c], Mahfoud Kadja[d]

[a]*LR3MI Laboratory, Mechanical Engineering Department, Faculty of Sciences for Engineering, BP 12, University of Annaba, Algeria*
[b]*Crystal Growth Laboratory, University of Victoria, Victoria, BC, Canada V8W3P6*
[c]*University of Lorraine, IUT de Longwy, 54400 Cosnes et Romain, France*
[d]*University of Constantine, Lab. Energétique & Pollution, Mech. Eng. Depart. Constantine, Algeria*

Abstract

Numerical simulations were carried out to explain the behavior exhibited in experimental work on the dissolution process of silicon into a germanium melt. The experimental work utilized a material configuration similar to that used in the Liquid Phase Diffusion (LPD) and Melt-Replenishment Czochralski (Cz) growth systems. The numerical simulations were carried out under the assumption of 2D. The mathematical model equations were developed using Lattice Boltzmann Method (namely the BGK approximation was adopted). Measured concentration profiles and dissolution height from the samples processed with and without the application of magnetic field show that the amount of silicon transported into the melt is slightly higher in the samples processed under magnetic field, and there is a difference in dissolution interface shape indicating a change in flow structure. This change in flow structure was predicted by the present LB model. In the absence of magnetic field a flat stable interface is observed. In the presence of an applied field, however, the dissolution interface remains flat in the center but curves back into the source material near the edge of the wall. This indicates a far higher dissolution rate at the edge of the silicon source.

Peer-review under responsibility of the scientific committee of ICOME 2015 and ICOME 2016.

Keywords: Lattice Boltzmann Method; Silicon-Germanium; Dissolution; Crystal Growth; Magnetic field

* Corresponding author. Tel.: +213-791-679-682; fax: +213-38875399
E-mail address: farid.mechighel@etu.unilim.fr

1876-6102
Peer-review under responsibility of the scientific committee of ICOME 2015 and ICOME 2016
10.1016/j.egypro.2017.11.188

1. Introduction

The lattice-Boltzmann method (LBM) provides an alternative to the conventional approach to computational fluid dynamics (CFD), in which the starting point is always a discretization of the Navier–Stokes equations. The method, which is basically based on Boltzmann's kinetic transport equation, instead describes a fluid by a number of interacting populations (called *distributions functions*) of particles moving and colliding on a fixed lattice. In recent years, the LBM has enjoyed much applied success modeling various complex flows in the domain of engineering interest (1–5].

In this paper we suggest a LBE model for modeling in crystal growth. In particular the configuration used in the model and simulation is the arrangement in which the silicon seed is floating on top of the germanium melt. In this case, the silicon seed covers the melt's free surface. A schematic of the material configuration used in this work is shown in figure 1. This arrangement is similar to the crucible stacking used in the liquid phase diffusion (LPD) growth system for SiGe single crystal [6–8].

In this work we carry out a numerical study for this configuration (under 2D assumption). Instead solving the usual (PDEs) equations (i.e. the Navier-Stokes equations and both the advection-diffusion equation for the energy balance and the species balance), the lattice Boltzmann technique considers and solves others equations derived from the *kinetic theory* of gases, which called *LBE "Lattice Boltzmann equations"* [1].

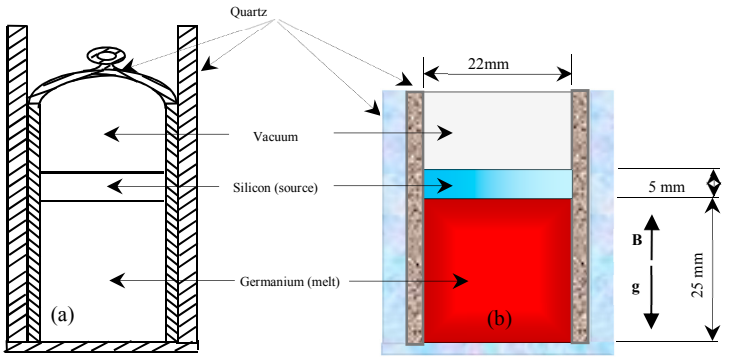

Figure 1. (a) The setup used in the experiment [9] and (b) the simulated domain [7].

2. Lattice Boltzmann Model

2.1. Lattice Boltzmann equation (LBE) for the melt flow

The lattice Boltzmann equation LBE for the melt flow is given as,

$$f_k\left(\mathbf{x}+\mathbf{c}_k, t+1\right)-f_k\left(\mathbf{x}, t\right)=\Omega_k+\mathbf{c}_k\cdot\mathbf{F}_k^{ext}, \tag{1}$$

with $k=0,\ 1,\ \ldots,\ q-1$ and where f_k is the one-particle distribution function associated with motion in the k^{th} direction. The term Ω_k of Eq. (1) is the *collision* operator that describes how the q values f_k defined on the same node at given time step interact [2, 3]. The populations can only move with a finite number of velocities $\mathbf{c}_k=\{c_0, c_1, \cdots c_{q-1}\}$. Furthermore, the model may be additionally simplified by specifying the collision integral as the Bhatnager - Gross - Krook (BGK) operator: $\Omega_k=\omega\left(f_k^{eq}-f_k\right)=\left(f_k^{eq}-f_k\right)/\tau$, where τ is the *relaxation* time. Now, equation (1) describes *streaming* dynamics together with relaxation to *local equilibrium*, f_k^{eq}, in time proportional to τ.

The macroscopic variables (for the hydrodynamic field): the velocity $\mathbf{u}$ and the density ρ are defined *locally* as *moments* of the distribution functions, such as,

$$\rho = \sum_{k=0}^{q-1} f_k \quad \text{and} \quad \rho \mathbf{u} = \sum_{k=0}^{q-1} \mathbf{c}_k f_k \,. \tag{2}$$

Furthermore the hydrodynamic pressure is given as: $p = \rho/3$.

The local equilibrium distribution functions, for Navier-Stokes equations, are taken as

$$f_k^{eq}(\mathbf{x},t) = \rho(\mathbf{x},t) w_k \left[1 + \frac{1}{c_s^2} \mathbf{c}_k \cdot \mathbf{u} + \frac{1}{2c_s^4} \mathbf{Q}_k : (\mathbf{u} \cdot \mathbf{u}) \right] \tag{3}$$

with the notation ($:$) is used for tensor product and the tensor $\mathbf{Q}_k$ is defined as follows: $\mathbf{Q}_k = \mathbf{c}_k \cdot \mathbf{c}_k + c_s^2 \mathbf{I}$. In equation (3) the constant c_s is the *speed of sound* of the model. This parameter, as well as, the q parameters 'w_k' are lattice constants. In practice, a value of $c_s^2 = 1/3$ is found to be numerically most *stable*, and this choice is therefore most commonly adopted [10, 11].

2.2. Lattice Boltzmann equations for heat and mass transports

Similar to momentum transport, the LB equations for energy and mass (solute) transports are given respectively as,

$$g_k(\mathbf{x}+\mathbf{c}_k, t+1) = g_k(\mathbf{x},t)[1-\omega_T] + \omega_T g_k^{eq}(\mathbf{x},t) \quad \text{and} \quad h_k(\mathbf{x}+\mathbf{c}_k, t+1) = h_k(\mathbf{x},t)[1-\omega_C] + \omega_C h_k^{eq}(\mathbf{x},t), \tag{4}$$

where ω_T and ω_C are the relaxation times towards equilibrium for energy and species transports respectively.

The macroscopic variables for energy and concentration equations are defined respectively as,

$$T(\mathbf{x},t) = \sum_{k=0}^{q-1} g_k \equiv \sum_{k=0}^{q-1} g_k^{eq} \quad \text{and} \quad C(\mathbf{x},t) = \sum_{k=0}^{q-1} h_k \equiv \sum_{k=0}^{q-1} h_k^{eq} \,. \tag{5}$$

Furthermore we use, respectively, the following equilibrium distribution functions for heat and species transports:

$$g_k^{eq}(\mathbf{x},t) = w_k T(\mathbf{x},t)\left[1 + (\mathbf{c}_k \cdot \mathbf{u})/c_s^2\right] \quad \text{and} \quad h_k^{eq}(\mathbf{x},t) = w_k C(\mathbf{x},t)\left[1 + (\mathbf{c}_k \cdot \mathbf{u})/c_s^2\right] \tag{6}$$

The relaxation times towards equilibrium for Navier-Stokes, energy and solute equations are given respectively as:

$$\omega = 1/\left(\upsilon^{LB}/c_s^2 + 0.5\right), \; \omega_T = 1/\left(\alpha^{LB}/c_s^2 + 0.5\right) \text{ and } \; \omega_C = 1/\left(D^{LB}/c_s^2 + 0.5\right). \tag{7}$$

2.3. Boundary conditions

Since it was assumed the *no-slip* condition for the fluid flow on the entire crucible walls, thus the *Bounce-back* conditions are imposed on the bottom, left, right and the top of the crucible. For the temperature field a Dirichlet boundary condition is imposed on the left, right and bottom surface, while on the top an adiabatic condition is adopted. For the concentration field all the crucible walls are insulated. At the dissolution interface the continuity condition is applied for velocity field.

Solid phase and dissolution interface

The problem can be greatly simplified when assuming that there is no species diffusion in the silicon solid, so that $\mathbf{u}_{solid} = \mathbf{0}$ (where $\mathbf{u}_{solid}$ is the velocity vector in the solid phase). A similar procedure may be applied for solid

phase by replacing the solid phase velocity ($\mathbf{u}_{solid} = \mathbf{0}$) in the previous system of equations. We get the LBE system of equations for the silicon solid phase (for more details see articles [12, 7].

3. Results and discussions

The simulations performed for the present crucible configuration, without magnetic field, exhibited an expected *diffusion-dominated* behavior in the dissolution process. Transport into the melt is relatively slow and continues to slow down as the concentration gradient flattens. The stable flow structure, caused by silicon buoyancy in the melt, results in a very flat dissolution interface. However, in the presence of an applied static magnetic field, the shape of the dissolution interface is significantly different as seen in figure 2. While the interface is flat everywhere under no magnetic field, with application of the magnetic field, areas near the crucible wall experience a higher dissolution rate and more material is removed into the melt [9, 7].

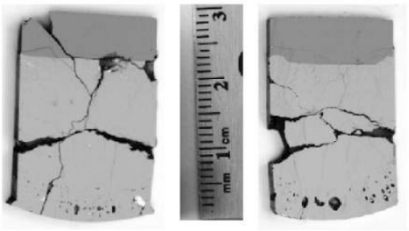

Figure 2. Experiment conducted with no field on the left and experiment conducted with field is on the right. The regions of high dissolution are easily visible under magnetic field. The pictures are taken from the reference [9].

The interface remains flat at the center of the material. However, near the wall the interface slightly curves into the material. There is more dissolution in this region than the center. This indicates a significant change to the melt flow structure. The upward strong hot convective flows, due to the action of combined thermosolutal buoyancy and magnetic body forces observed in the crucible near the heated lateral surface, hit the interface at the edges and contribute to the solute transport near this region. Some of these hot upward flows turn away to the center of the crucible and hence bring the diffused solute away from the edge to the center of the bulk melt. In the region close to the center of the interface, the convective flow is very weak [9, 7]. This is due to the domination of the diffusion in this region. The flow structures under various magnetic fields are illustrated in figures 3 and 5 (for lack of space only the 0.8 Tesla field is presented).

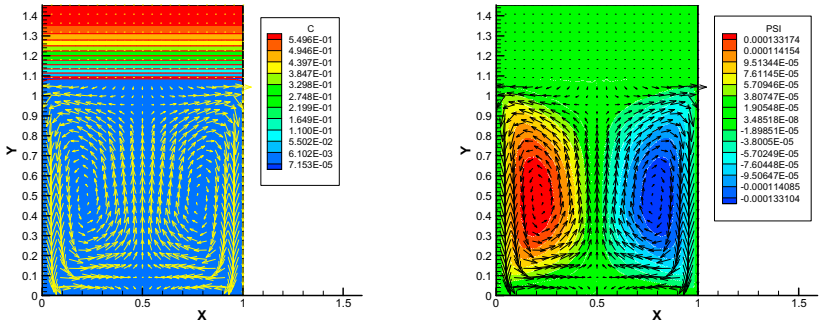

Figure 3. Simulation with no applied field. Arrows indicate flow structure, and isolines illustrate concentration profile. The profile around the dissolution interface is flat.

The magnetic field appears to be acting to mix silicon away from the crucible wall and into the center. This action creates a higher concentration gradient at the crucible edge with increasing dissolution. Due to a slight increase in dissolved silicon, it appears that the applied field does not have a significant effect on the vertical (y-

direction) flow structure, this is well observed in the distribution of the vertical velocity components, while the horizontal (x-direction) flow component appear to be increased (Figs. 5 and 6). These observations were supported by experiments [9, 13]. Note that here we have performed the simulation using a simplified 2D model, for which tri-dimensionality is ignored. The impact of the tri-dimensionality of the flow on the interface shape was well emphasized experimentally by [9] and numerically in [7].

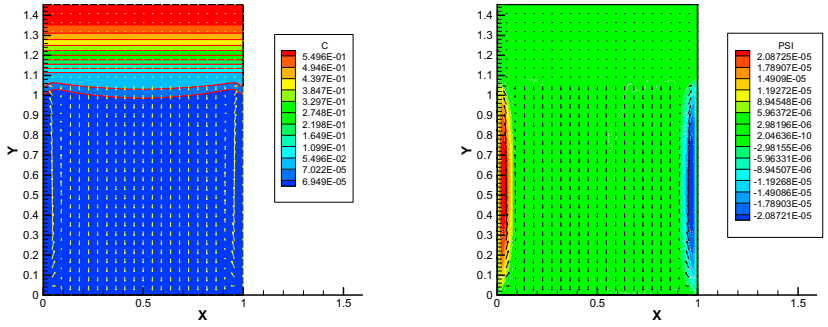

Figure 4. Simulation with 0.8 Tesla applied field. Arrows indicate flow structure, and isolines illustrate concentration profile. The profile around the dissolution shows that silicon is being mixed away from the crucible edge towards the center.

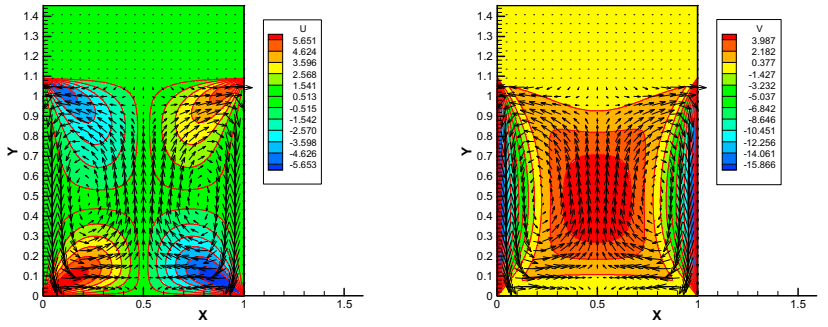

Figure 5. Simulation with no applied field. Arrows indicate flow structure, and isolines illustrate horizontal/vertical velocity component profile. Note that U and V are given in LB system of units.

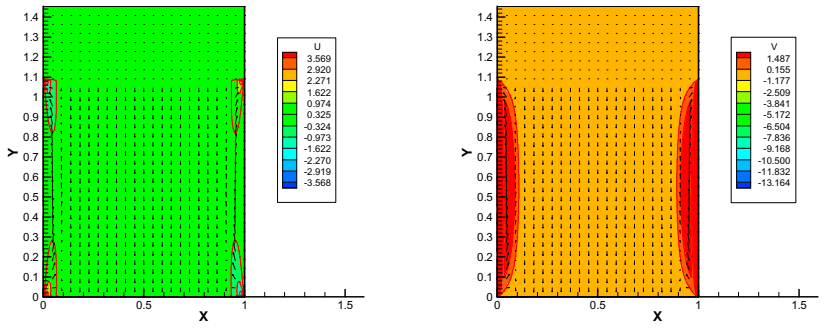

Figure 6. Simulation with 0.8 Tesla applied field. Arrows indicate flow structure, and isolines illustrate horizontal/vertical velocity component profile. Note that U and V are given in LB system of units.

It is well known that a static magnetic field is frequently utilized to suppress thermosolutal convection in crystal growth. In the present crucible configuration, it appears that the flow is not suppressed. Instead, the applied magnetic field enhances the already weak stable flow structure. Indeed, the applied field strengthens the upward flow near the lateral heated crucible wall and damps the downward flow in the crucible core. An external magnetic field, aligned perfectly with the axis of the growth cell (y-direction in our present 2D model), gives rise to a magnetic body force in the horizontal plane that balances the vertical gravitational body force, and consequently may weaken the convective flow [9, 7].

4. Conclusions

The LBM numerical simulations conducted, using a 2D model, lead to the following conclusions:

- Transport in a silicon germanium system, where silicon is being dissolved from the top of the melt, exhibits a *diffusion*-dominated behaviour.
- The application of a static magnetic field to the melt does not reduce the strength of the flow structure (near the lateral heated crucible wall). As the system is already relatively free of thermosolutal convection, the applied magnetic field serves to strengthen the *vertical* flow component. This appears to cause significant *mixing* of silicon away from the crucible wall into the core of the melt. This phenomenon may have application in controlling growth interface geometry. Specifically, it may be utilized to maintain a constant growth interface curvature during the LPD growth cycle [9, 7].
- The numerical results of the present setup show that the silicon dissolution was slightly enhanced under an applied vertical magnetic field. This enhancement peaked for field levels between 0.3 to 0.5 Tesla. The magnetic level of 0.8 Tesla is a fit choice for obtaining uniform concentration distribution. This observation can be attributed to the altered flow structure in the melt due to the magnetic field.
- Finally by comparing numerical results and experiments it appears that the present 2D model may predict the flow structure and the interface shape, but the impact of the tri-dimensionality of the flow on the dissolution shape should be elucidated. Thus three dimensional simulations are required for the present growth system.

References

[1] Guo Z, and Shu C. Lattice Boltzmann Method and Its Applications in Engineering, World Scientific Publishing Company; 1st ed.; 2013
[2] Mohamad AA. Applied Lattice Boltzmann Method for Transport Phenomena, Momentum, Heat and Mass Transfer, Sure Print, Dalbrent, Calgary, Canada; 2007
[3] Succi S. The Lattice Boltzmann Equation for Fluid Dynamics and Beyond, Clarendon press, Oxford; 2001
[4] Chen S, Doolen GD. Lattice Boltzmann Method for Fluid Flows. Annu. Rev. Fluid Mech. 1998; 30: 329–364.
[5] Lallemand P, Luo, LS. Theory of Lattice Boltzmann Method: Acoustic and Thermal Properties in Two and Three Dimensions, Phys. Rev. E 2003; 68:036706-1–036706- 25.
[6] Yildiz M, Dost S, Lent B. [2005], Growth of bulk SiGe single crystals by liquid phase diffusion, J. Crystal Growth 2005; 280:151–160.
[7] Mechighel F, Armour N, Dost S, Kadja M. Axisymmetric and 3-D Numerical Simulations of the Effects of a Static Magnetic Field on Dissolution of Silicon into Germanium, CMES: Computer Modeling in Engineering & Sciences 2014; 97(1):53-80.
[8] Yildiz M. A Combined Experimental and Modeling Study for the Growth of SixGe1-x Single Crystals by Liquid Phase Diffusion (LPD), PhD thesis, University of Victoria, Canada; 2005.
[9] Armour N, Dost S. Effect of an applied static magnetic field on silicon dissolution into a germanium melt. J. Crystal Growth 2009; 311(3):780-782.
[10] Qian Y. Simulating thermohydrodynamics with Lattice BGK Models, J. Sci. Comput. 1993;8:231–242.
[11] Lätt J. Hydrodynamic Limit of Lattice Boltzmann Equations, PhD thesis, University of Geneva; 2007.
[12] Mechighel F, Armour N, Dost S, Kadja M. Mathematical modeling of the dissolution process of silicon into germanium melt. TWMS: J. App. Eng. Math. 2011, 1(2):127-149.
[13] Armour N. Transport Phenomena in Liquid Phase Diffusion Growth of Silicon Germanium, PhD thesis, Univ. of Victoria, Canada; 2012.