NUMERICAL MODELLING OF BORON REMOVAL FROM SILICON WITH OXIDIZING GAS JET

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Abstract: One of the most perspective methods to produce solar grade silicon is refinement via metallurgical route. The most critical part of this route is refinement from boron and phosphorus. One possible approach to remove boron is use of reactive gas on surface of silicon melt.

This paper focuses on 2D and 3D numerical analysis of dynamics of boron removal from fixed gas-silicon interface (chemical reactions are not taken into account) in order to estimate mass transfer coefficient of boron and to study its dependence on geometry and placement of lance(s) of oxidizing gas injecting system.

Key words: boron removal, mass transfer coefficient, oxidizing gas, silicon refinement

1. Boron mass transfer coefficient at gas-silicon interface. Molten silicon refining by means of boron removal with incoming gas is long-term process with duration of several hours.

Efficiency of boron removal can be characterized with boron mass transfer coefficient k_B at gas–silicon interface, which can be estimated using the dependence of boron concentration $C_{B,t}$ in silicon on refining process time t [1]

$$ln\frac{C_{B,t}}{C_{B,0}} = -k_B\frac{S_{g|Si}}{V_{Si}}t \tag{1}$$

where $C_{B,0}$ is boron concentration at refining process beginning $(t = 0)$, $S_{g|Si}$ is area of gas–silicon interface, V_{Si} is silicon volume.

2. Computational model. Dynamics of long-term silicon refinement is studied based on developed 2D and 3D models (see Figures 1 and 4 (a,c,e)) with the following domains:

- *silicon*, which has been melt in cylindrical crucible by alternating current in ring coil (crucible and coil are not shown in schemes)
- *gas* is located over molten silicon
- gas jet is supplied over single (multiple) *lance(s).*

Physical parameters of considered models are collected in Tables 1.

In reality, during refinement process, there is a chemical reaction taking action on the gas–silicon interface surface: reacting gas oxidizes boron and new molecules are formed which carry boron away from the melt.

In described simulation an idealized system is used, where single scalar variable, t.i. concentration of boron, is used in both domains – in molten silicon and in gas.

In the beginning of refinement process the fixed concentration of boron in melt is assumed. Then, gas, which is injected over lance without boron (t.i. boron concentration is equal to zero) is blown on the surface of silicon, thus part of boron is removed.

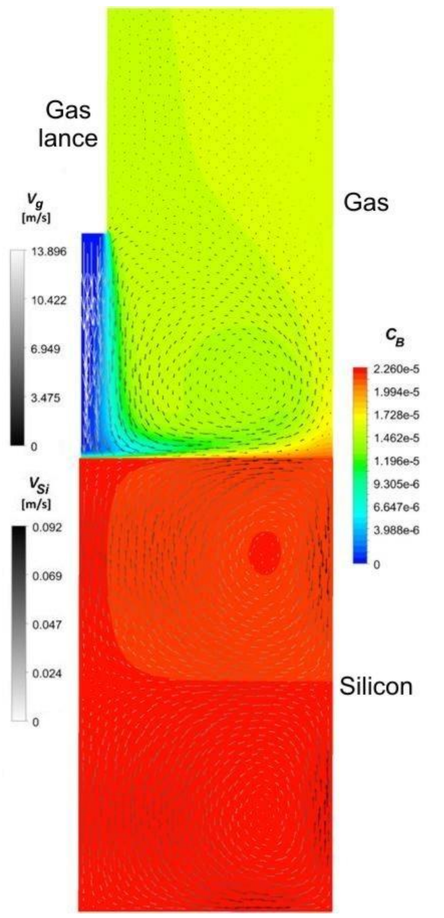

Figure 1: 2D scheme of boron removal from silicon with gas jet: v_{Si} and v_g – silicon and gas velocity vectors; C_B – contours of boron concentration, computes with 2D model for flow time $t = 1$ *h*.

Geometry parameters:
Silicon or gas radius – 0.031 m
Silicon or gas height – 0.062 *m*
Radius of gas jet lance – 0.0031 *m*
Distance of lance from gas-silicon interface – 0.031 *m*

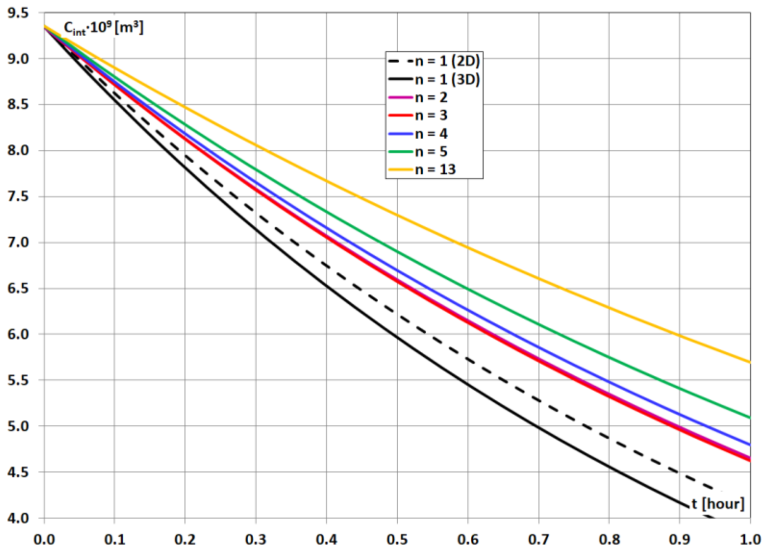

Figure 2: Boron integral concentration C_{int} in silicon melt as function of silicon refining process time *t* for different numbers (*n*) of vertical ($\alpha_{lance} = 0°$) gas lances with jet velocity $v_g = 13.53$ *m/s*

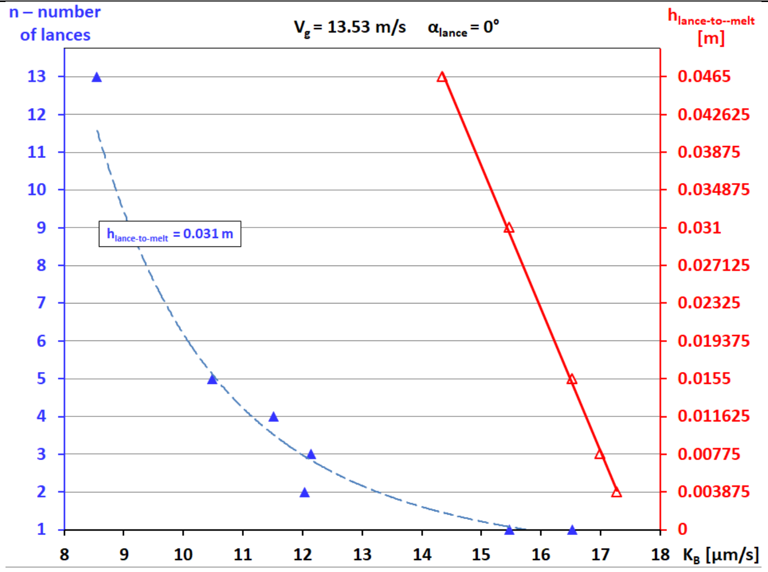

Figure 3: Boron mass transfer coefficient k_B at gas-silicon interface for vertical ($\alpha_{lance} = 0°$) gas lances with jet velocity $v_g = 13.53$ *m/s*:
(red scale and triangles) k_B as function of gas lance height $h_{lance\text{-}to\text{-}melt}$ with linear trend line (solid)
(blue scale and triangles) k_B as function of number of lances *n* for $h_{lance\text{-}to\text{-}melt} = 0.031$ *m* with power trend line (dotted)

Table 1: Parameters of computational model

Gas	Silicon (*Si*)
Temperature – 1400˚C Density – 0.38932 kg/m^3 Dynamic viscosity – $6.542\cdot 10^{-5}$ $kg/(m\cdot s)$ Flow rate at normal conditions – 4 l_N/min Inlet velocity – 13.53 *m/s*	Temperature – 1426.85˚C Density – 2547 kg/m^3 Dynamic viscosity – $6.05\cdot 10^{-4}$ $kg/(m\cdot s)$
	Boron (*B*)
	Diffusivity in gas $3.4\cdot 10^{-4}$ m^2/s Diffusivity in silicon $2.4\cdot 10^{-8}$ m^2/s

Quasi-stationary gas and silicon flow patterns and boron concentration distribution is shown on Figure 1. Distributions of 2D fields in Figure 1 are similar to computed distributions for free chosen vertical cross-section of axis-symmetrical 3D model.

3. Computational procedure. Dynamics of long-term refinement process of molten silicon by means of boron removal with gas jet is modelled with ANSYS software.

- The computations of silicon melt flow in crucible and gas flow injected with lance are performed using ANSYS FLUENT package with application of k–ε turbulence model.

Boundary conditions for velocity at gas–silicon interface are the following: no slip conditions for gas and free slip conditions for silicon. These conditions are the first approximation of coupled boundary conditions for shear stresses in the case of extremely small value of gas viscosity η_g in comparison with silicon viscosity η_{Si}

- Boron concentration both in melt and in gas is modelled with UDS (User Defined Scalar) option in ANSYS FLUENT with application of coupled boundary conditions, which present conservation conditions of boron mass flux

$$D_{B,Si}\frac{\partial C_{B,Si}}{\partial n} = D_{B,g}\frac{\partial C_{B,g}}{\partial n} \qquad (2)$$

where $C_{B,Si}$ and $C_{B,g}$ are boron concentrations at gas–silicon interface in silicon and gas accordingly as well as $D_{B,Si}$ and $D_{B,g}$ are diffusivity of boron in silicon melt and gas.

- Lorentz force, induced by coil, which is driver of silicon induction stirring, is modelled with EMAG module of ANSYS Multiphysics or with ANSYS Maxwell packages.

4. Peculiarities of numerical procedure. In order to save computational resources in multi-variants research of boron refinement process the series of physical fields' transient analysis have been performed with 2D and axis-symmetrical 3D models:

- determination of flow time value, which corresponds to quasi-stationary state of gas and silicon melt flows;
- determination of time step values with equivalent time dependence of integral concentration of boron in silicon melt for fixed gas and silicon flow patterns, which correspond to quasi-stationary state of flows.

The further computations have been performed with the following conditions:

- Gas and melt flows and B concentration transient analysis is carried out with time step $\tau \sim 0.001$ s till quasi-stationary state of flows, t.i. flow time value $t \sim 15–25$ s.
- Transient analysis of B concentration with fixed gas and silicon flows patterns is carried out with time step value $\tau \sim 1$ s.

5. Comparison of computational and experimental results. Computed integral boron concentration in silicon during long-time refinement process is shown in Figure 2 – dotted and solid lines for 2D and axis-symmetrical 3D models with single lance (n = 1) accordingly.

The estimated boron mass transfer coefficient values based on simulation of one hour refinement is k_B = 14.04 *μm/s* for 2D model; for axis-symmetrical 3D model estimated value is for 10% greater – k_B = 15.46 *μm/s* (see Figure 2). These values are in good agreement with measurement results in [2, 3].

6. Computational results for boron mass transfer coefficient at gas-silicon interface. In order to estimate efficiency of long-time boron refinement process the dependence of mass transfer coefficient of boron on geometry and placement of lance(s) of gas injecting system is studied.

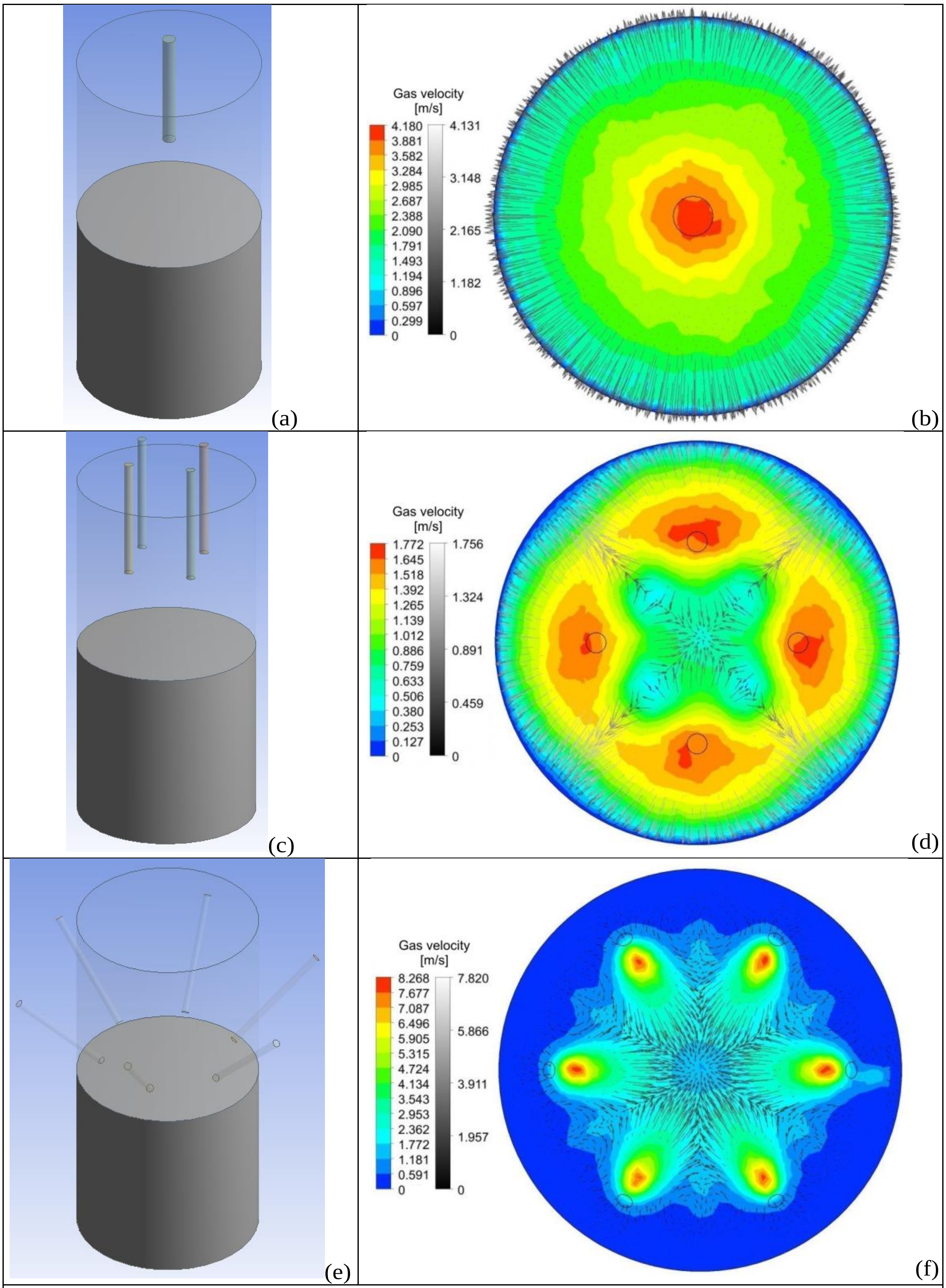

Figure 4: (a) Axis-symmetrical and (c,e) rotational-symmetrical 3D models with silicon (lower region), gas (upper region) and different numbers (n): (a) $n = 1$; (c) $n = 4$ – or (e) $n = 6$.
(a,c) Vertical ($\alpha_{lance} = 0°$) or slanted ($\alpha_{lance} = 45°$) gas lances.
Gas lance height $h_{lance\text{-}to\text{-}melt}$: (a,c) 0.031 m, (e) 0.00775 m.
(b,d,f) Gas velocity contours (color scale) and vectors (inverse grey scale) in horizontal cross-section with distance 0.0651 m from silicon–gas interface for corresponding geometry (a,c,e).

6.1. Distance of inlet lance from melt top surface for single vertical lance. Coefficient k_B as function of gas lance height $h_{lance\text{-}to\text{-}melt}$ is shown in Figure 3 (red scale and triangles) for single vertical ($\alpha_{lance} = 0°$) gas lances with constant jet velocity $v_g = 13.53$ *m/s*.

Obtained results can be approximated with linear trend line (see solid line in Figure 3).

6.2. Different number (n) of vertical lances ($\alpha_{lance} = 0°$). The computations are performed for $n = 1, 2, 3, 4, 5, 13$. Several geometries are shown in Figure 4 (a,c). Gas jet velocity is constant $v_g = 13.53$ *m/s*. Total flow rate of all gas jets in scope of chosen model is constant and is equal to flow rate of gas jet for model with single lance.

Coefficient k_B as function of number of lances n for $h_{lance\text{-}to\text{-}melt} = 0.031$ *m* is shown in Figure 3 (blue scale and triangles). Obtained results, which can be approximated with power trend line (dotted line), show, that coefficient k_B decreases in accordance with increase of number of lances, where the largest value of k_B corresponds to single lance system.

Possible explanation is concerned with analysis of gas flow patterns in vicinity of gas–silicon interface – see Figure 4 (b,d) with velocity field distribution in horizontal cross-section with distance 0.0651 *m* from silicon–gas interface:

- Every gas jet, which is supplied over the lances pool, is weaker in mass flow rate if compare with gas jet supplied over single lance (conservation of total gas flow rate is taken into account).
- Gas jets decelerate each other and break gas return flow from silicon surface to outlet in the top of model – see velocity contours and vectors in Figure 4 (b,d).

6.3. Slanted lances. Obtained results show, that coefficient k_B decreases in accordance with increase of angle of slope α_{lance} which is counted from vertical axis of gas lance.

For model with changed direction of gas return flow – along the axis of model (see Figure 4 (e,f)) versus along the side surface of crucible (see Figures 4 (c,d)) – mass transfer remain less efficient in comparison with one single lance model (see Figures 4 (a,b)).

7. Conclusions. Two- and three-dimensional numerical models are developed for analysis of dynamics of boron removal from fixed gas-silicon interface (chemical reactions are not taken into account). The aim is estimation of mass transfer coefficient of boron for widely varying physical and geometrical parameter of system including placement and spatial orientation of lance(s) of gas injecting system.

Computational results show that the most effective is axis-symmetrical gas-injecting system with single lance, placed as close to silicon surface as possible – these systems are characterized with greater values of mass transfer coefficient at gas–silicon interface.

Acknowledgment. This work was funded by European Regional Development Fund under contract “Refinement of metallurgical grade silicon using smart refinement technologies” (No. 1.1.1.1/16/A/097).

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