

DOI: 10.1515/amm-2016-0090

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## NUMERICAL MODELING OF COPPER REDUCTION IN FIRE REFINING PROCESS

Copper reduction represents one of the steps in the process of fire refining of blister copper. Process of copper reduction with natural gas as performed in the anode furnace was modeled. Numerical analysis was conducted using the IPSA (Inter-Phase-Slip Algorithm) module of PHOENICS package and standard turbulent model  $k - \epsilon$ . The model takes into account the liquid phase - slag and phase of natural gas which is supplied through the tuyere from the bottom of the charge. Calculations were made for two different diameters of tuyeres for supply of reducer. Based on the calculations and analysis, it was found out that the gas flow which causes strong movement in the bath guarantees homogeneous composition of the liquid copper in each process stage. Application of a tuyere with a larger diameter results in a greater intensity of the reduction process and better use of the reducer.

*Keywords:* numerical modelling, copper reduction, anode furnace, tuyere

### 1. Introduction

The development in computing technology resulted in application of engineering numerical methods in many fields. Using the methods of CFD (Computational Fluid Dynamics) it is possible to optimize processes by changing their characteristics.

In the literature there are many studies into numerical modeling of processes in converters. The dynamics of flow in the Teniente converter was analyzed with physical model and numerical modeling. Identification of flow ranges was made by changing the Froude number, and the frequency of bath movement as a function of its height and depth of tuyere immersion was presented [1,2]. In other study [3], process of fluid flow and beginning of turbulences in the converted copper was physically and numerically modeled for different velocities of the gas flow. Mechanism of formation, growth and detachment of gas bubbles was observed.

In the presented study, a numerical model of the process of copper reduction with natural gas was made based on the phenomenon of two-phase flow of liquid-gas mixture. Bubbling phenomenon represents a specific example of such a flow. It consists in the flow of the gas phase bubbles through the liquid phase layer [4,5]. The conducted numerical analysis provided possibilities to determine progress of the reduction process, distribution of velocity of both phases, as well as distribution of oxygen concentration versus time during the reduction process. The analysis was performed for two diameters of tuyeres which supply the reducer to the bath.

Reduction of copper is one of the steps in the whole process of fire refining. Fire refining process is carried out in the anode rotary furnaces. The furnaces have a capacity of 250 - 270 Mg of liquid copper.

The charge to the rotary kilns is in a form of liquid blister copper from flash-smelting furnace and converter copper from the refining of Cu-Pb-Fe alloy. The reduction process is carried out by blowing of natural gas into the charge through tuyeres which are located at the bottom of the furnace. The copper temperature at the end of the reduction stage is 1200-1220°C. The final oxygen concentration in the copper after reduction is up to 0.12 wt.%. [6,7].

### 2. Geometrical and mathematical model

The geometric scope of the model covers that part of the furnace which is filled with liquid metal. Because of the symmetry of the furnace along the vertical plane, which is perpendicular to the furnace axis, only half of the furnace has to be taken into consideration in the calculations.

The geometry of the developed model of anode furnace in which the reduction process was carried out consisted of the following elements:

- copper charge
- tuyere – natural gas inlet
- outlet of gases – top copper surface.

The copper charge was contained in a rectangular block of dimensions representing half of the amount of the processed charge of 130 Mg. At the bottom the inlet of the tuyere of diameter 0.038 m was located for introduction of the natural gas.

For the calculations, the following assumptions were made:

- liquid copper of following properties:

- temperature 1200°C
- density 7687 kg/m<sup>3</sup>
- thermal conductivity 170 W/m K

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- kinetic viscosity  $4.72 \times 10^{-7} \text{ m}^2/\text{s}$
- specific heat capacity  $495 \text{ J/kg K}$
- nitrogen-rich natural gas of following properties:
  - temperature  $20^\circ\text{C}$
  - density  $0.8 \text{ kg/m}^3$
  - thermal conductivity  $0.016 \text{ W/m K}$
  - kinetic viscosity  $1.53 \times 10^{-5} \text{ m}^2/\text{s}$
  - specific heat capacity  $1902 \text{ J/kg K}$
  - calorific value  $29.6 \text{ MJ/Nm}^3$

and simplified chemical composition:

- CH<sub>4</sub> 80 vol. %.
- N<sub>2</sub> 20 vol. %.

The calculations were made in two variants, with a change of the diameter of the tuyere which supplies gas in an amount of  $400 \text{ Nm}^3/\text{h}$ :

1.  $d = 38 \text{ mm}$  (diameter of the tuyere used in industrial tests)
2.  $d = 54 \text{ mm}$

In the second case the cross-sectional area of the tuyere was two times larger. To maintain the same flow of the reducer two times lower gas inlet velocity was declared. In the variants 1 and 2 the velocity was  $100 \text{ Nm/s}$  and  $50 \text{ Nm/s}$ , respectively.

The analyzed domain was of the following dimensions:  $4.7 \times 2.4 \times 1.4 \text{ m}$ . The structural grid as generated in Phoenixics program consisted of 22 000 differential elements [8].

In the study it was assumed that adiabatic flow takes place, i.e. without any heat exchange processes. Due to the heterogeneity of phase distribution it was necessary to use multiphase models. In addressing the two-phase flow problem the IPSEA (Inter-Phase-Slip Algorithm) method was used, in which the Navier-Stokes equations are solved for each phase separately [9,10].

In description of the turbulent flows  $k - \varepsilon$  model was used. This model is based on two equations which describe the turbulent kinetic energy  $k$  and dissipation rate  $\varepsilon$  [11]. The following equations define the parameters:

$$\frac{\partial}{\partial t}(\bar{\rho} k) + (\bar{\rho} k \bar{u}_i)_{,i} = (\bar{\tau}_{ij} \bar{u}_{j,i}) - \bar{\rho} \varepsilon + (\mu_k k_{,i})_{,i} \quad (1)$$

$$\frac{\partial}{\partial t}(\bar{\rho} \varepsilon) + (\bar{\rho} \varepsilon \bar{u}_i)_{,i} = c_{\varepsilon 1}(\bar{\tau}_{ij} \bar{u}_{j,i}) - \frac{c_{\varepsilon 2} \bar{\rho} \varepsilon^2}{k} + (\mu_\varepsilon \varepsilon_{,i})_{,i} \quad (2)$$

where:

$t$  – time (s),

$\rho$  – density of fluid ( $\text{kg m}^{-3}$ ),

$k$  – turbulent kinetic energy ( $\text{m}^2 \text{ s}^{-2}$ ),

$\varepsilon$  – dissipation rate of turbulent kinetic energy ( $\text{m s}^{-3}$ ),

$u$  – vector of flow velocity ( $\text{m s}^{-1}$ ),

$\tau_{ij}$  – turbulent stress tensor ( $\text{N m}^{-3}$ ),

$c_{\varepsilon 1}, c_{\varepsilon 2}, \sigma_k, \sigma_\varepsilon, c_\mu$  – constants,

$$\mu_k = \mu + \frac{\mu_T}{\sigma_k}$$

$$\mu_\varepsilon = \mu + \frac{\mu_T}{\sigma_\varepsilon}$$

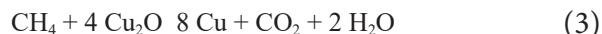
$$\mu_T = \rho c_\mu k^2 / \varepsilon$$

$\mu$  – molecular viscosity ( $\text{kg m}^{-1} \text{ s}^{-1}$ ),

$\mu_T$  – turbulent viscosity ( $\text{kg m}^{-1} \text{ s}^{-1}$ ).

Reduction is a periodical process of changing with time parameters, therefore the calculation was performed for the transient system.

Natural gas is supplied through the tuyere and flows through liquid copper in a form of bubbles. The reduction process follows the reaction Eq. (3) and takes place at the boundary of gaseous and liquid phases.



The reduction process can be simply considered as the transfer of oxygen from the liquid copper to the gas bubble. The amount of oxygen  $\Delta m_{O_2}$  which is transferred in the specific time of the reduction process from the liquid copper can be described by the following relation - Eq. (4).

$$\Delta m_{O_2} = F \times a \times \Delta \tau \quad (4)$$

where:

$F$  – surface area of mass exchange,

$a$  – oxygen transfer coefficient,

$\Delta \tau$  – time step.

In order to determine the average coefficient of oxygen transfer the data from an industrial plant were used. For the selected copper reduction cycle in a furnace the following parameters were determined:

- volume of gas bubble  $2.87 \times 10^{-5} \text{ m}^3$
- surface area of gas bubble  $4.5 \times 10^{-3} \text{ m}^2$
- velocity of bubble outflow  $0.43 \text{ m/s}$
- time of bubble flow through liquid layer  $3.2 \text{ s}$
- total number of bubbles generated in reduction cycle  $335 \times 10^5$
- surface of interphase contact in reduction cycle  $167\,000 \text{ m}^2$

The average oxygen transfer coefficient  $a_{av}$  was determined from Eq. (5), where  $m_{Cu}$  represents the amount of processes copper, while  $g'_{O_2}$  and  $g''_{O_2}$  concentration of oxygen in the copper before and after the reduction process, respectively. "A" is the surface of interphase of all gas bubbles, and  $\tau$  is the time of contact of bubbles with liquid copper.

$$a_{av} = \frac{m_{Cu} \times (g'_{O_2} - g''_{O_2})}{A \times \tau} \quad (5)$$

Thus determined the average oxygen transmission coefficient amounts to  $3.27 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ .

The value of this coefficient depends on the mass concentration of oxygen in copper and mass concentration of methane in the gaseous phase. In order to take those relationships into account it was assumed that the oxygen transfer coefficient is a linear function of oxygen concentration in copper as well as a linear function of methane concentration in the gas. After appropriate transformations the following relationship was established - Eq. (6):

$$\Delta m_{O_2} = 26,7 a_{av} \sqrt{g_{O_2l} \times g_{CH_4g}} \quad (6)$$

where:

$g_{O_2l}$  – mass concentration of oxygen in liquid phase,

$g_{CH_4g}$  – mass concentration of methane in the gaseous phase.

After replacing the „ $a_{av}$ ” with actual data, the function as defined by Eq. (7) was produced.

$$\Delta m_{O_2} = 0,0873 \sqrt{g_{O_2l} \times g_{CH_4g}} \quad (7)$$

The surface area of mass transfer was determined after taking the assumption that in the average differential element there are bubbles of uniform size (according to the theory of bubbling van Krevelen). Considering the diversity of the dimensions of the generated MR mesh and the volume fraction of gas  $R_g$  in the differential element the mass transfer surface area was described by Eq. (8).

$$F = 132 (1-R_g) R_g (V_{ER})^{2/3} \quad (8)$$

where:

$V_{ER}$  – volume of differential element expressed in  $m^3$ .

Masses of the components of liquid (Cu and  $O_2$ ) and gaseous ( $CH_4$ ,  $CO_2$ ,  $H_2O$  and  $N_2$ ) phases in the differential elements were determined using Eq. (9) and Eq. (10).

$$m_{comp\ g} = V_{ER} R_g \rho_g g_{comp\ g} \quad (9)$$

$$m_{comp\ l} = V_{ER} R_l \rho_l g_{comp\ l} \quad (10)$$

where:

$R_g, R_l$  – volume fraction of the gaseous phase and liquid phase in the differential element,

$\rho_g, \rho_l$  – density of gaseous and liquid phase,

$g_{comp\ g}, g_{comp\ l}$  – mass concentrations of components of gaseous and liquid phase.

After a specified time step the amount of components in the differential elements will change as a result of the reduction processes - Eq. (11).

$$g_{comp\ l,g} = \frac{m_{comp\ l,g} + \Delta m_{comp\ l,g}}{\Sigma_{comp} m_{comp\ l,g}} \quad (11)$$

where:

$m_{comp\ l,g}$  – amount of component of liquid or gaseous phase in ER,

$\Delta m_{comp\ l,g}$  – increase of the amount of component of liquid or gaseous phase in the time step  $\Delta t$ .

Concentrations of the components after the time step will amount to:

$$g_{comp\ l,g} = \frac{m'_{comp\ l,g}}{m'_{l,g}} \quad (12)$$

### 3. Results of model calculations

Following are the results of model calculations for the variant 1, i.e. a tuyere with diameter of 38 mm, through which the gas is supplied in the amount of 400  $Nm^3/h$ , which corresponds to the 100  $Nm/s$  velocity of the gas outlet from the tuyere.

Distribution of oxygen concentration in the vertical section of the furnace along the axis of the furnace after 30, 60, 90 and 120 minutes, are shown in Fig. 1 - 4. Uniform field of oxygen concentration was obtained in each of the illustrated stages of reduction.

TABLE 1 lists the values of oxygen concentration for a central differential element of the domain in question as well as the minimum and maximum values in the whole studied area. It can be seen that as the duration of the reduction process increases, the oxygen concentration in the copper decreases to reach the value of 0.1% after 120 minutes.

TABLE 1  
Oxygen concentration in copper [%]

Reduction time	In central ER	Lowest	Highest
30 min.	0.5547	0.5431	0.5664
60 min.	0.3245	0.3208	0.3282
90 min.	0.1888	0.1878	0.1898
120 min.	0.1098	0.1095	0.1101

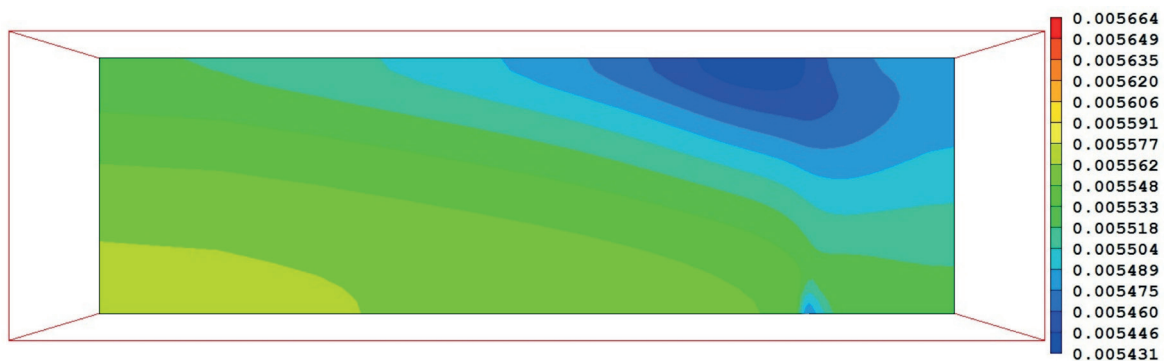


Fig. 1. Oxygen concentration distribution in copper after 30 min

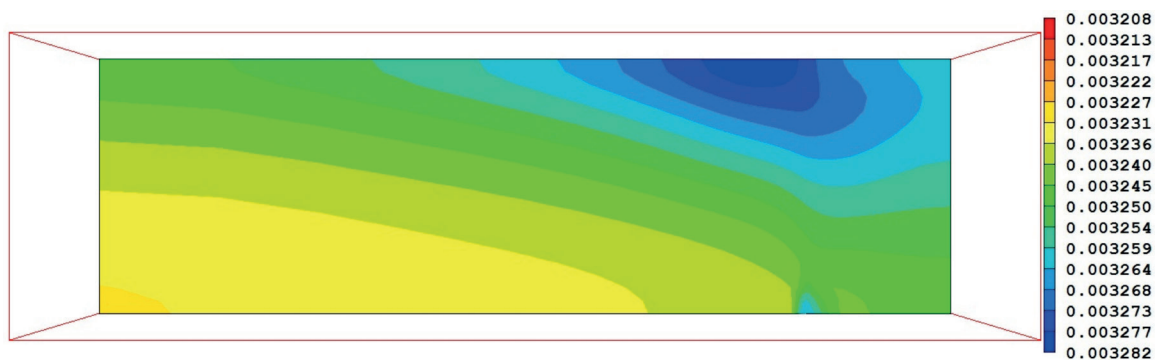


Fig. 2. Oxygen concentration distribution in copper after 60 min

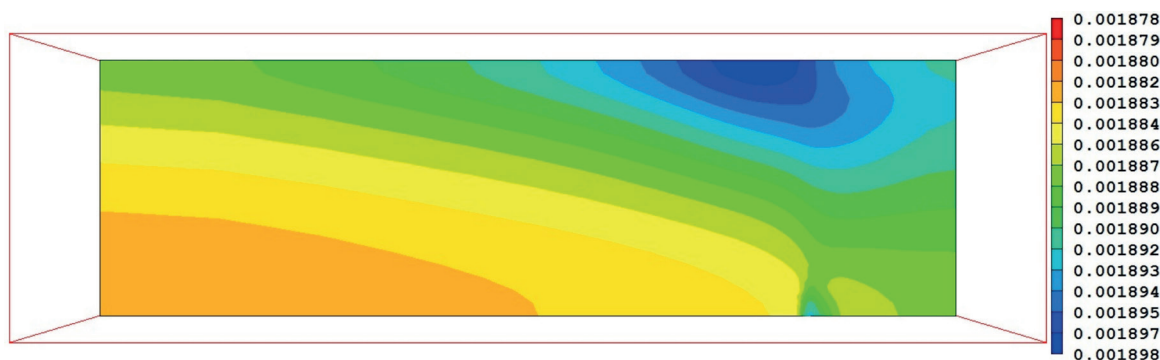


Fig. 3. Oxygen concentration distribution in copper after 90 min

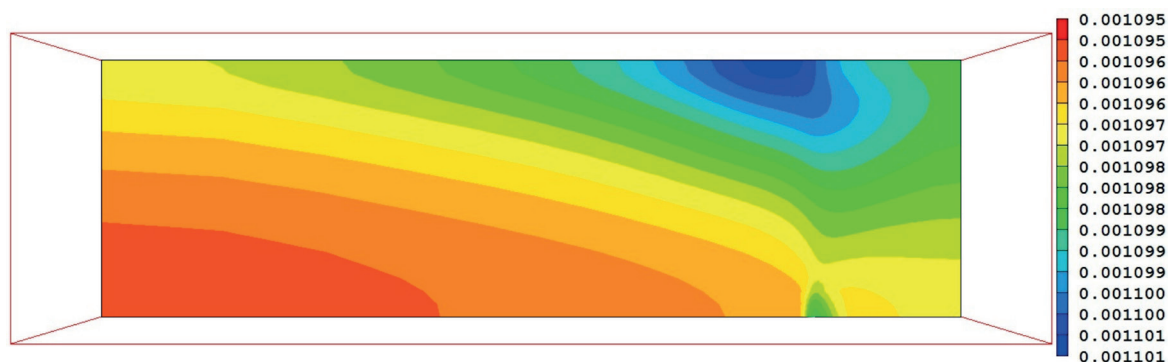


Fig. 4. Oxygen concentration distribution in copper at the end of reduction process

Uniformity of the oxygen concentration field results from the intensity of mixing of liquid copper with the flowing gases. The gaseous phase moves upward by the action of buoyancy force and it is carried by the liquid phase.

The distribution of its velocity is shown in Fig. 5. The velocity values are presented by the color scale while the vectors refer to the liquid phase. Formation of rotary motion in the liquid which is stirred by the gaseous phase can be observed.

The distribution of the gaseous phase in the vertical plane is shown in Fig. 6.

In the next part of the paper results of calculations produced for the second variant with application of a tuyere of larger diameter - 54 mm and the velocity of gas at the tuyere outlet of 50 Nm/s are presented.

The distribution of oxygen concentration in copper which was reached after two hours of reduction process is in the range  $0.0648 \div 0.0687\%$  and is lower than for the variant 1 tuyere (Fig. 7).

Fig. 8 shows progress of reduction process as determined for the tuyeres of two different diameters. Comparison of the progresses shows that in the first 40 minutes the reduction process with the tuyere of 38 mm diameter is more intense. After 40 minutes application of a tuyere with a larger diameter results in more intense reduction. With the larger tuyere better use of the reducer is achieved. This is not, of course, a general conclusion but related to the modeled system, defined tuyere diameters and flow parameters.

Justification for this result can be observed in making comparison of the velocity distributions and distributions



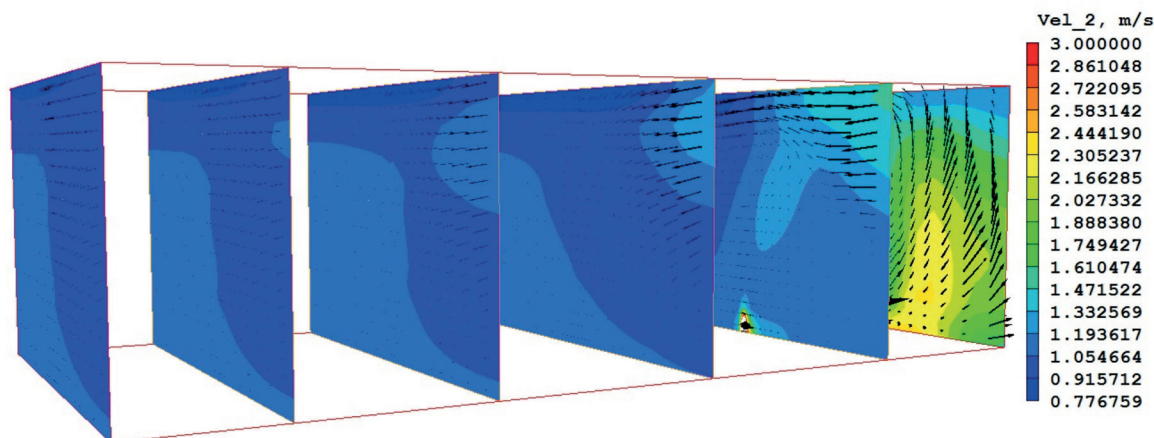


Fig. 5. Distribution of gaseous phase velocity



Fig. 6. Share of the gaseous phase in vertical section

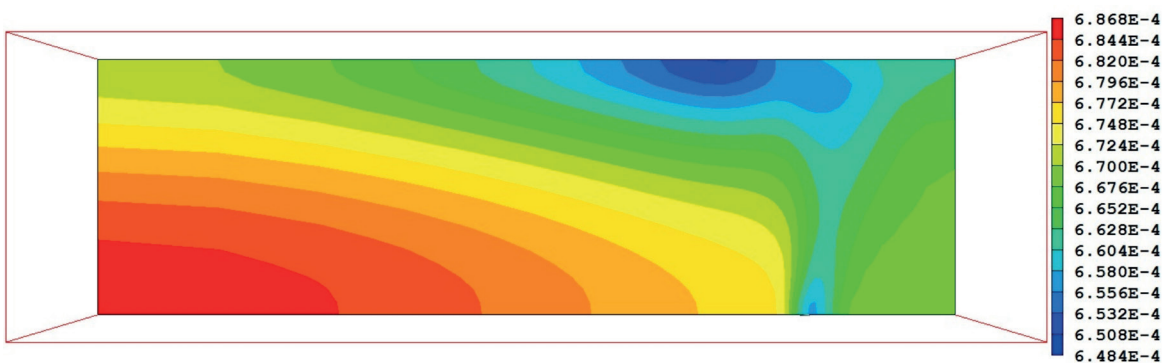


Fig. 7. Oxygen concentration distribution in copper at the end of reduction process (variant 2)

of a gaseous phase ( $R_g$ ) in the considered variant. Velocity distributions in planes perpendicular to the axis of the furnace are shown in Fig. 9. As in the previous variant, the velocity distributions in the transverse sections of the furnace are more uniform; the furnace space is better used when a tuyere with a larger diameter is applied.

Comparison of the shares of the gaseous phase in the variant 1 (Fig. 6), and in the variant 2 (Fig. 10) shows presence of the gaseous phase in the wider area in the variant 2.

When taking reduction process progress as a decisive

criterion the superiority of the tuyere of larger diameter was demonstrated. In practice, the physical phenomenon of tuyere fouling and the associated need for its periodic clearing takes place. Therefore, there are two opposing factors to be considered. On one hand, larger diameter of the tuyere brings better utilization of the reducer and intensifies the process, on the other hand it increases the tendency for tuyere fouling. In this case, relevant studies have to be carried out to determine the optimal parameters of the tuyere.

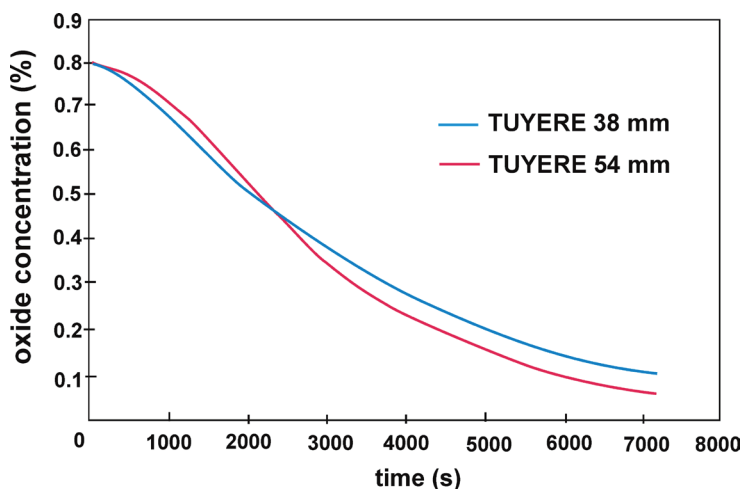


Fig. 8. Change in oxygen concentration in copper during reduction process

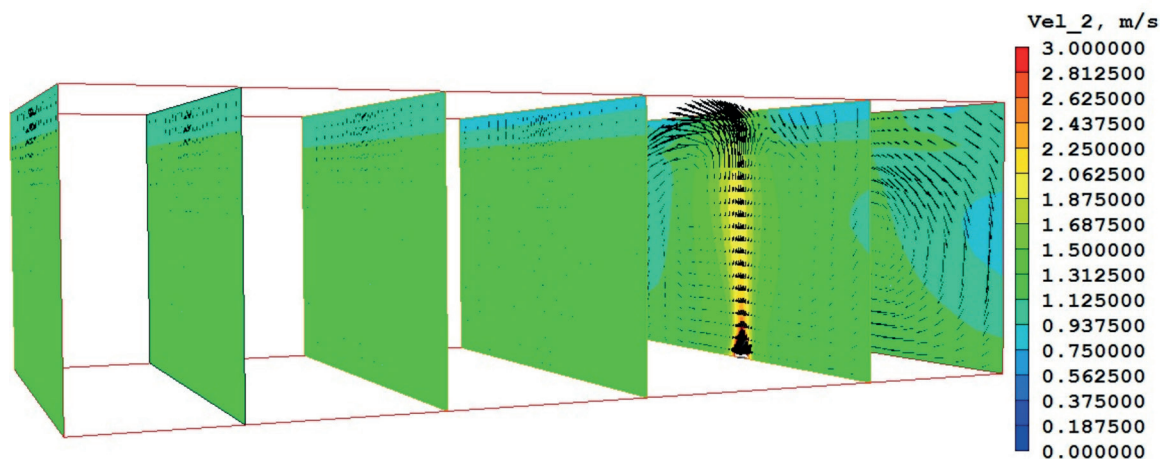


Fig. 9. Distribution of gaseous phase velocity (tuyere diameter 54 mm)

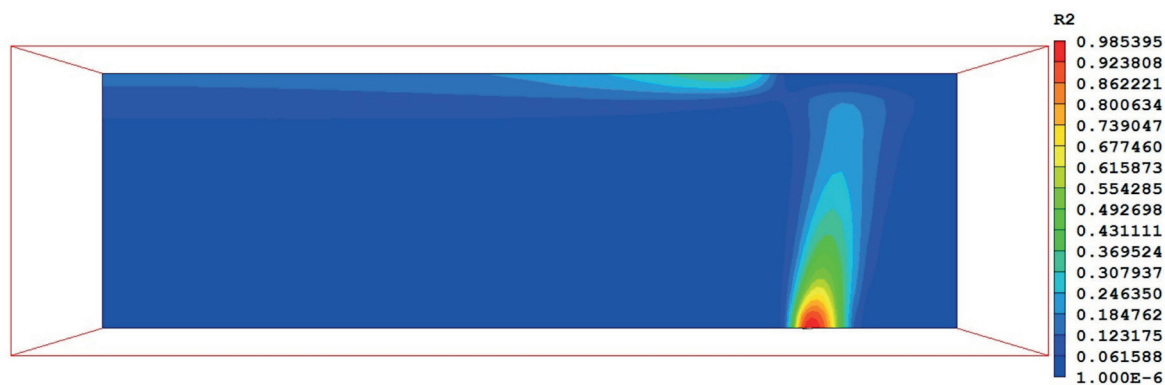


Fig. 10. Share of the gaseous phase in horizontal section at the top surface of the melt (variant 2)

#### 4. Conclusions

Numerical calculations were made for two variants, each with different size of the gas supplying tuyere. The results of calculations led to the following conclusions:

The intense movement of the bath caused by the gas flow results in homogeneous liquid copper composition in each process step. The dispersion of oxygen concentration in the liquid copper does not exceed  $\pm 3\%$  and decreases with the

progress of reduction.

The model calculations show the superiority of the tuyere with a diameter of 54 mm above the tuyere with a diameter of 38 mm, when the reduction process intensity is considered. Application of a larger tuyere results in a better use of the reducer and acceleration of the reduction process.

To optimize the selection of tuyeres it is necessary to perform experiments in industrial facility because of the simplifications of any mathematical model.

### Acknowledgements

Studies were conducted within the scope of the project “New technologies and new construction of machines and equipment for enrichment and metallurgical treatment of mineral resources” no POIG.01.03.01-24-019/08 co-financed by the European Union from Regional Development Fund.

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