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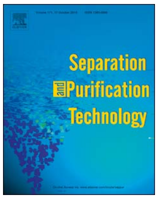

Effect of off-centered silicon ladle on the removal strength of aluminum and calcium impurities

Jie Wang[a,b], Zhengjie Chen[a,b,*], Wenhui Ma[a,b,*], Kuixian Wei[a,b], Shaoyuan Li[b], Junru Liu[a,b], Weimin Ding[c], Jianhua Wen[a,b]

[a] *Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming 650093, China*
[b] *State Key Laboratory of Complex Nonferrous Metal Resources Cleaning Utilization in Yunnan Province, Kunming University of Science and Technology, Kunming 650093, China*
[c] *Nujiang Hongsheng Kam Silicon Limited, Nujiang 673201, China*

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ABSTRACT

The bottom blowing gas in the silicon ladle plays an essential role in the purification of industrial silicon, and nozzle arrangement has an important effect on silicon quality after refinement. In the present study, a mathematical model has been proposed to assess the transient three-dimensional and two-phase flow in rich oxygen bottom-blowing silicon ladle with a single centered and off-centered porous nozzle, respectively. The numerical simulation is performed with the multiphase flow model of volume of fluid to model gas blown through a ladle. Fluid flow characteristics and the dead zone proportion between the centered and off-centered ladle are compared. The results demonstrate that the flow performance of liquid-silicon is better, agitation kinetics are more robust, and the dead zone area is smaller in bottom blowing of the off-centered ladle than of the centered ladle, which aligns well with the results of industrial experiments. The amount of Al and Ca impurities in silicon decreases by 13.22% and 25.87%, respectively, and the impurity distribution is more uniform.

1. Introduction

Silicon is widely used in metallurgy, electronics, and the chemical industry. With the rapid development of the polysilicon [1] and photovoltaic fields, demand has increased regarding silicon product quality. Silicon purification is therefore a topic of interest for researchers. Extensive work has been done to obtain high-quality silicon products via secondary refinement [2–4], acid leaching [5,6], vacuum purification [7], directional solidification [8,9], plasma melting [10], and electron beam melting [11]. Among these, ladle metallurgy presents an economical and practical means of refining industrial silicon. It is widely used in industry [12–14] to effectively remove harmful impurity elements, homogenize temperature and chemical compositions, and improve liquid silicon quality.

The nozzle position, number of nozzles, and gas flow rate must be considered when refining bottom-blowing technology in silicon ladles. During practical operation, occasionally the gas flow is too weak to facilitate sufficient reactions between gas and impurities. A high gas flow rate returns the slag from the melt surface to the liquid silicon and may result in secondary pollution. Improper nozzle arrangement leads to unsatisfactory impurity removal. It is difficult to obtain the characteristics of fluid flow and the contact area between gas and molten silicon due to the high operating temperature and lack of appropriate detection methods. Fortunately, mathematical modeling of gas-liquid flow [15] for metallurgical applications is highly developed, especially in the iron and steel industries. Many studies [16–24] focused on critical phenomena in gas-stirred ladles with different types of nozzles in recent years. For example, Zhu et al. [25] developed a water model and a three-dimensional mathematical model to investigate the flow feature of liquid steel and mixing phenomena in argon gas bottom-stirring ladles with six types of tuyere arrangements. They found that the shortest mixing occurred in a single tuyere in an off-center position, whereas the opposite placement of double tuyeres at half radii was the best configuration considering the aspects of blowing, inclusion flotation, mixing, and splashing. Liu et al. [26] conducted a numerical simulation to describe the quasi-steady liquid steel flow and interfacial behavior of a three-phase argon-stirred ladle with one and two off-centered plugs placed at 180° and 90° arrangements. The volume of fluid (VOF) model was used to capture the free surface of immiscible phases. They found that the proper selection of argon flow rate and plug configuration during ladle refinement depended on different refining purposes including mixing time and metallurgical reactions. Torres

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* Corresponding authors at: Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming 650093, China.
E-mail addresses: czjkmust@126.com (Z. Chen), mwhsilicon@126.com (W. Ma).

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et al. [27] investigated the stirring of an argon injection steel ladle with a top and bottom nozzle using computational fluid dynamic simulations. Three top-submerged lance depths and three argon gas flow rates were considered; the stirring phenomenon and mixing time were discussed in the numerical experiments. Cloete et al. [28] applied a full-scale numerical model to gas-stirred ladles to analyze the relationship between multiple design variables and mixing efficiency by employing the discrete phase and VOF models. Ladle height was the most significant factor for mixing efficiency, and gas injection through multi-tuyere was also a promising strategy that relied largely on tuyere arrangements to improve mixing. Zhang et al. [29] researched fluid flow characteristics, mixing time, and interfacial behavior between slag and steel in an argon bottom-injection ladle with a diverse off-centered porous plug at different gas flow rates using a transient and three-phase flow numerical model.

Numerical simulation is an effective auxiliary method to analyze gas-liquid multiphase flow in the metallurgical industry. However, few studies have concentrated on optimizing the silicon ladle to improve silicon purity by adopting a mathematical model. In this work, a mathematical model is developed to research the transient three-dimensional and gas-liquid flows in rich oxygen bottom-blowing silicon ladle with a single centered and off-centered porous nozzle. A special gas flow rate is largely determined by the actual temperature, and the weight of molten silicon is employed in the numerical simulation. The VOF model is used to analyze the behavior of the gas-liquid interface. Comparisons of the characteristics of fluid flow, turbulent kinetic energy, and the dead zone area between the centered and off-centered ladle are conducted to strengthen refinement. At the same time, the existence of metal impurities (Fe, Al, and Ca) seriously influences silicon performance, use, and value. It is difficult to remove Fe via a conventional method given its lower affinity for oxygen compared to silicon. Some industrial trials have been conducted with an off-centered silicon ladle to further remove Al and Ca impurities; findings concur with the numerical simulation results.

2. Mathematical model

2.1. Model assumptions

Some reasonable assumptions were employed in the mathematical model for silicon ladle fluid flow, as shown below:

(1) In the calculation process, chemical reactions were not considered, the energy equation was not applied, and the effect of temperature on the gas and liquid phases was ignored.
(2) The initial height of the static molten silicon was 1140 mm, the influence of slag was neglected, and the silicon in the ladle was considered an incompressible fluid.
(3) The interface between gas and liquid silicon was calculated by the VOF model, and interfacial tensions were considered.
(4) Non-slipping conditions were used for all solid walls, and standard wall functions were applied at nodes near the wall.

2.2. Basic equations

Liquid silicon, oxygenized air, and atmospheric air were taken into account in the numerical simulation. The fluid flow and turbulent performance were controlled by the continuity equation, the momentum equation (Navier-Stokes equations), k-ε turbulence model, and VOF model. The equations are as follows:

(1) Continuity equation

$$\frac{\partial \rho}{\partial t} + \nabla\cdot(\rho\vec{v}) = 0 \tag{1}$$

(2) Momentum equation

$$\frac{\partial}{\partial t}(\rho\vec{v}) + \nabla\cdot(\rho\vec{v}\vec{v}) = -\nabla p + \nabla\cdot[\mu_e(\nabla\vec{v} + \nabla\vec{v^T})] + \rho\vec{g} + F \tag{2}$$

where ρ, v, and p represent the fluid density, velocity, and pressure, respectively; g is the acceleration of gravity; and μ_e is the effective turbulent viscosity. The effective turbulent viscosity is calculated by the k-ε turbulence model, which can be written as follows:

(3) The k-ε turbulence model

$$\mu_e = \mu_t + \mu = \rho C_\mu \frac{k^2}{\varepsilon} + \mu \tag{3}$$

$$\rho\left(\frac{\partial k}{\partial t} + \frac{\partial k\mu_i}{\partial x_i}\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\partial_k}\right)\frac{\partial k}{\partial x_j}\right] + G_k - \rho\varepsilon \tag{4}$$

$$\rho\left(\frac{\partial \varepsilon}{\partial t} + \frac{\partial \varepsilon\mu_i}{\partial x_i}\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\partial_\varepsilon}\right)\frac{\partial \varepsilon}{\partial x_j}\right] + C_{1\varepsilon}\frac{\varepsilon}{k}G_k - C_{2\varepsilon}\rho\frac{\varepsilon^2}{k} \tag{5}$$

where k is turbulent kinetic energy, ε is the turbulent dissipation rate, and G_k is the generation of turbulent kinetic energy due to the mean velocity gradients. C_u, $C_{1\varepsilon}$, and$C_{2\varepsilon}$ are constants; $C_u = 0.09$, $C_{1\varepsilon} = 1.44$, and $C_{2\varepsilon} = 1.92$.

(4) Volume of fluid model

$$\frac{\partial a_q}{\partial t} + \vec{v}\cdot\nabla a_q = 0 \tag{6}$$

The VOF model depends on two or more phases that are not interpenetrating. q represents the different phases, and a_q is the phases' volume fraction.

2.3. Modeling Geometry and conditions

Fig. 1 shows the prototype's geometric dimensions of a silicon ladle with an off-centered porous nozzle. A three-dimensional numerical model was developed based on the prototype. The calculation domain grid consisted of 330,000 cells, and the inlet area grid was refined. Entry boundary conditions and outlet boundary conditions were set to the velocity inlet and pressure outlet, respectively. The velocity of oxygenized air was calculated based on actual air flow. Fluid properties of the phases and other operating parameters are shown in Table 1.

The three-dimensional unsteady state and Pressure Implicit with Splitting of Operator (PISO) algorithm were used for the simulated calculation. Second Order Upwind was adopted for the momentum equation, and Implicit Body Force was employed to improve the convergence of results. The mathematical model solution was calculated via Fluent 15.0 simulation software.

3. Industrial trials

The silicon ladle with a centered nozzle is often used in factories. A new optimized ladle, which was identical the original except for the nozzle being located at one-third radii (1/3 r) was designed based on numerical simulation, as shown in Fig. 1. During actual production, oxygenized air was injected through a porous nozzle at the bottom of the ladle to purify the liquid silicon. To ensure accuracy of the results, primitive and optimized ladles were used to refine the molten silicon of the same batch from the submerged arc furnace. Five sets of industrial experiments were completed with the centered and off-centered ladle. Relevant experimental phenomena and data were recorded during the refinement process. After refining the centered and off-centered ladle, the contents of aluminum and calcium impurities were analyzed and compared. The distribution of impurities in different parts of the silicon ingot was investigated when the molten silicon cooled into the ingot. The respective effects of the primitive and optimized ladles on