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Mathematical model of multistage and multiphase chemical reactions in flash furnace^①

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Abstract: A mathematical model of multistage and multiphase reactions in flash smelting furnace, which based on the description of chemical reactions and reaction rate, is presented. In this model, main components of copper concentrate are represented as FeS₂ and CuFeS based on experiment, intermediate products are assumed to be S₂ and FeS, and the final products are assumed as FeS, FeO, SO₂, Cu₂S, FeO and FeO(SiO₂)₂. The model incorporates the transport of momentum, heat and mass, reaction kinetics between gas and particles, and reactions between gas and gas. The *k-ε* model is used to describe gas phase turbulence. The model uses the Eulerian approach for the gas flow equations and the Lagrangian approach for the particles. The coupling of gas and particle equations is performed through the particle source in cell (PSIC) method. Comparison between the model predictions and the plant measurements shows that the model has high reliability and accuracy.

Key words: flash smelting furnace; mathematical model; copper; simulation; CFD

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1 INTRODUCTION

The flash smelting process is widely used throughout the world for copper production, accounting for about 50% of global capacity for primary copper production. It is based on the utilization of the heat released by oxidation reactions of the sulfuric concentrates. The feed mixture in flash smelting process is fed through the concentrate burner into a reaction shaft. Then the concentrate particles react with gas phase to a predetermined oxidation degree. After the reaction shaft, the particle-gas suspension flows to the settler, where the particles (drops) separate from the gas and form slag and matte layers on the bottom of the settler.

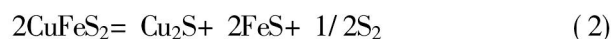
Numerical modeling of fluid field, temperature field and concentration field in flash smelting furnace make it possible for production enhancement, energy saving and increasing the lifetime of concentrate burner and reaction shaft^[1-3]. During 1980s, Themelis^[4] and Sohn^[5-9] made valuable 1-D or 2-D modeling efforts on mathematical modeling of fluid flow and heat transfer in a flash smelting furnace shaft. Recently, many modeling studies have been performed in Commonwealth Scientific and Industrial Research Organization, Australia and Outokumpu^[10-14]. These studies had too many assumptions and still kept in laboratory research stage, and none of them passed the plant test and used to direct or optimize

the production.

2 CONCENTRATE MULTISTAGE REACTION MODEL

The heat sources in flash smelting furnace are the oxidation reactions of iron and sulfur in concentrates. Most researchers took the reactions in flash smelting furnace as one-step reaction. This assumption could simplify programming, but it caused that the temperature of particles were higher than the real temperature and the accuracy of model was reduced. This paper used three stages to describe reactions in flash smelting furnace.

1) The decomposition reactions occur after the particle's temperature reaches the decomposition one. The decomposition reactions are endothermic and produce the intermediate products, namely FeS and S₂:



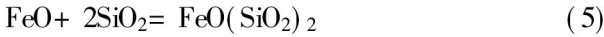
2) The labile sulfur from the chalcopyrite and pyrite combusts with the oxygen and releases heat to the gas phase. Partial FeS combusts with the oxygen and products FeO and SO₂. At the same time, reaction releases heat to the particles directly:



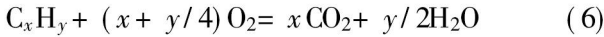
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3) Slag blow reaction:



Oil is necessary when the combustion heat of concentrate could not meet the need of the flash smelting process:



The thermal decomposition rate of sulfur from concentrate is computed from the following expression:

$$R_{S, \text{div}} = R_{S, \text{FeS}_2} + R_{S, \text{CuFeS}_2} \\ = \alpha_1 \pi d_p^2 \rho_p k_{\text{dis}, \text{FeS}_2} + \alpha_2 \pi d_p^2 \rho_p k_{\text{dis}, \text{CuFeS}_2} \quad (7)$$

where R_{S, FeS_2} and R_{S, CuFeS_2} are decomposition rates of FeS_2 and CuFeS_2 , α_1 and α_2 are masses of labile sulphur per kilogram sulphide in FeS_2 and CuFeS_2 , d_p is the particle diameter, ρ_p is the density of particle.

$k_{\text{dis}, \text{FeS}_2}$ and $k_{\text{dis}, \text{CuFeS}_2}$ are overall rate coefficients for decomposition reactions of S_2 from FeS_2 and CuFeS_2 :

$$\frac{1}{k_{\text{dis}, \text{FeS}_2}} = \frac{1}{k_{S, d}} + \frac{d_p(d_p - d_{c1})}{2d_{c1}D_S} + \frac{d_p^2}{d_{c1}^2 k_{S, \text{FeS}_2}} \quad (8)$$

$$\frac{1}{k_{\text{dis}, \text{CuFeS}_2}} = \frac{1}{k_{S, d}} + \frac{d_p(d_p - d_{c1})}{2d_{c1}D_S} + \frac{d_p^2}{d_{c1}^2 k_{S, \text{CuFeS}_2}} \quad (9)$$

where $k_{S, d}$ is mass transfer coefficient of sulphur, D_S is diffusivity of sulphur, k_{S, FeS_2} and k_{S, CuFeS_2} are decomposition rate coefficients of labile sulphur in FeS_2 and CuFeS_2 .

These rates are computed with Arrhenius. d_{c1} is a diameter of an unreacted chalcopyrite or pyrite core, as shown in Fig. 1. $k_{S, d}$ is estimated from the following correlation:

$$Sh = 2 + 0.6Re^{1/2}Sc^{1/3} \quad (10)$$

where Sh is Sherwood number, Sc is Schmidt number, Re is particle Reynolds number.

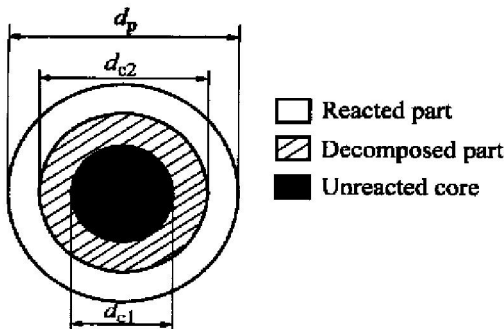


Fig. 1 Particle reaction core model

D_S can be calculated by the following equation:

$$D_S = \frac{1.01 \times 10^{-12} T^{1.75} \left[\frac{1}{W_S} + \frac{1}{W_{N_2}} \right]^{1/2}}{p \left[\left(\sum V_S \right)^{1/3} + \left(\sum V_{N_2} \right)^{1/3} \right]^2} \quad (11)$$

where W_S and W_{N_2} are relative molecular mass of sul-

phur and nitrogen, $\sum V_S$ and $\sum V_{N_2}$ are critical volume of sulphur and nitrogen in standard boiling point, p is pressure.

R_{ox} is the oxygen consumption rate of FeS:

$$R_{\text{ox}} = \pi d_p^2 \rho_p k_{\text{ox}, \text{ov}} Y_{\text{O}_2} \quad (12)$$

$k_{\text{ox}, \text{ov}}$ is overall rate coefficient for oxidant reaction of FeS:

$$\frac{1}{k_{\text{ox}, \text{ov}}} = \frac{1}{k_{\text{ox}, d}} + \frac{d_p(d_p - d_{c2})}{2d_{c2}D_{\text{ox}}} + \frac{d_p^2}{d_{c2}^2 k_{\text{ox}, \text{dis}}} \quad (13)$$

where $k_{\text{ox}, d}$ is mass transfer coefficient of oxygen and estimated from Eqn. (10), D_{ox} is diffusivity of oxygen, $k_{\text{ox}, \text{dis}}$ are decomposition rate coefficients of oxygen, d_{c2} is diameter of thermally decomposed sulphide.

D_{ox} can be calculated by

$$D_{\text{ox}} = \frac{1.01 \times 10^{-12} T^{1.75} \left[\frac{1}{W_{\text{O}_2}} + \frac{1}{W_{N_2}} \right]^{1/2}}{p \left[\left(\sum V_{\text{O}_2} \right)^{1/3} + \left(\sum V_{N_2} \right)^{1/3} \right]^2} \quad (14)$$

where W_{O_2} is relative molecular mass of oxygen, $\sum V_{\text{O}_2}$ is critical volume of oxygen in standard boiling point.

3 GAS PHASE EQUATIONS

The gas phase is viewed from Eulerian framework. The steady state equations take the following general form:

$$\frac{\partial}{\partial x} (\rho_g u_g \phi) + \frac{\partial}{\partial y} (\rho_g v_g \phi) + \frac{\partial}{\partial z} (\rho_g w_g \phi) - \\ \frac{\partial}{\partial x} (\Gamma_\phi \frac{\partial \phi}{\partial x}) - \frac{\partial}{\partial y} (\Gamma_\phi \frac{\partial \phi}{\partial y}) - \frac{\partial}{\partial z} (\Gamma_\phi \frac{\partial \phi}{\partial z}) = S^\phi \quad (15)$$

where ρ_g is gas density, Γ_ϕ is exchange coefficient for variable ϕ , S^ϕ is source or sink term, ϕ is variable. Table 1 shows all the gas phase governing equations. The standard k - ϵ model was used to represent gas-phase turbulence.

$$G = \mu_1 \left\{ 2 \left[\left(\frac{\partial u_g}{\partial x} \right)^2 + \left(\frac{\partial v_g}{\partial y} \right)^2 + \left(\frac{\partial w_g}{\partial z} \right)^2 \right] + \left[\frac{\partial u_g}{\partial x} + \frac{\partial v_g}{\partial y} \right]^2 + \left[\frac{\partial u_g}{\partial x} + \frac{\partial w_g}{\partial z} \right]^2 + \left[\frac{\partial v_g}{\partial y} + \frac{\partial w_g}{\partial z} \right]^2 \right\} \quad (16)$$

$$\mu_\epsilon = \mu_1 + \mu_2 \quad (17)$$

$$\mu_1 = C_\mu \rho_g k^2 / \epsilon \quad (18)$$

Gas combustion used Eddy Break-up model and the sources was computed using the following equations:

$$S_{\text{g}^{\text{S}_2}}^Y = - \rho_g \frac{\epsilon}{k} 23.6 \left[\frac{\mu_g \epsilon}{\rho k^2} \right]^{1/4} \cdot \\ \min \left[Y_{\text{S}_2}, \frac{Y_{\text{O}_2}}{2 \frac{W_{\text{O}_2}}{W_{\text{S}_2}}}, \frac{Y_{\text{SO}_2}}{\left[1 + 2 \frac{W_{\text{O}_2}}{W_{\text{S}_2}} \right]} \right] \quad (19)$$

$$S_{\text{g}^{\text{SO}_2}}^Y = - 2 \frac{W_{\text{SO}_2}}{W_{\text{S}_2}} S_{\text{g}^{\text{S}_2}}^Y \quad (20)$$

Table 1 Gas phase equations

Equation	ϕ	Γ_ϕ	S^ϕ
Continuity	1	0	S_p^m
x momentum	u_g	μ_e	$-\frac{\partial p}{\partial x} + \frac{\partial}{\partial x}(\mu_e \frac{\partial u_g}{\partial x}) + \frac{\partial}{\partial y}(\mu_e \frac{\partial v_g}{\partial x}) + \frac{\partial}{\partial z}(\mu_e \frac{\partial w_g}{\partial x}) - \frac{2}{3} \frac{\partial}{\partial x}(\rho_g k) + \rho_g g_x + S_p^u + S_g^u$
y momentum	v_g	μ_e	$-\frac{\partial p}{\partial y} + \frac{\partial}{\partial x}(\mu_e \frac{\partial u_g}{\partial y}) + \frac{\partial}{\partial y}(\mu_e \frac{\partial v_g}{\partial y}) + \frac{\partial}{\partial z}(\mu_e \frac{\partial w_g}{\partial y}) - \frac{2}{3} \frac{\partial}{\partial y}(\rho_g k) + \rho_g g_y + S_p^v + S_g^v$
z momentum	w_g	μ_e	$-\frac{\partial p}{\partial z} + \frac{\partial}{\partial x}(\mu_e \frac{\partial u_g}{\partial z}) + \frac{\partial}{\partial y}(\mu_e \frac{\partial v_g}{\partial z}) + \frac{\partial}{\partial z}(\mu_e \frac{\partial w_g}{\partial z}) - \frac{2}{3} \frac{\partial}{\partial z}(\rho_g k) + \rho_g g_z + S_p^w + S_g^w$
Turbulent kinetic energy	k	$\frac{\mu_c}{\sigma_k}$	$G - \rho_g \varepsilon$
Dissipation rate	ε	$\frac{\mu_c}{\sigma_\varepsilon}$	$(\varepsilon/k)(C_1 G - C_2 \rho_g \varepsilon)$
Gas mass fraction Y_i	Y_i	$\frac{\mu_c}{\sigma_Y}$	$S_p^Y + S_g^Y$
Enthalpy	h_g	$\frac{\mu_c}{\sigma_h}$	$Q_{ip}^h + Q_{rg}^h + u_g \frac{\partial p}{\partial x} + v_g \frac{\partial p}{\partial y} + w_g \frac{\partial p}{\partial z} + S_p^h + S_g^h$

u_g, v_g, w_g are velocities in direction x, y and z ; g_x, g_y, g_z are gravity acceleration in direction x, y and z ; μ_e, μ_t, μ_l are effective gas viscosity, turbulence viscosity and laminar viscosity; S_p^i, S_g^i are particle source term and gas source term gotten from equation; σ is turbulent Prandtl number; i represent the $S_2, O_2, SO_2, C_xH_y, H_2O, CO_2$ in gas mass fraction Y_i .

$$S_{g^{C_xH_y}}^Y = -\rho_g \frac{\varepsilon}{k} 23.6 \left[\frac{\mu_c \varepsilon}{\rho_g k^2} \right]^{1/4} \cdot \min \left[Y_{C_xH_y}, \frac{Y_{O_2}}{(x+y/4) \frac{W_{O_2}}{W_{C_xH_y}}} \right] \quad (21)$$

$$S_{g^{CO_2}}^Y = -x \frac{W_{CO_2}}{W_{C_xH_y}} S_{g^{C_xH_y}}^Y \quad (22)$$

$$S_{g^{H_2O}}^Y = -\frac{y W_{H_2O}}{2 W_{C_xH_y}} S_{g^{C_xH_y}}^Y \quad (23)$$

$$S_{g^{O_2}}^Y = (x+y/4) \frac{W_{O_2}}{W_{C_xH_y}} S_{g^{C_xH_y}}^Y + 2 \frac{W_{O_2}}{W_{S_2}} S_{g^{S_2}}^Y \quad (24)$$

4 PARTICLE PHASE EQUATION

The particle phase is viewed from a Lagrangian framework. The first goal of the Lagrangian approach is to locate the particles along their trajectory in the flow field. This is performed by solving the particle equation of motion.

$$m_p \frac{dV_p}{dt} = C_D \rho_g |\mathbf{u}_a - \mathbf{v}_p| (\mathbf{u}_a - \mathbf{v}_p) \frac{A_d}{2} + m_p \mathbf{g} \quad (25)$$

where m_p is mass of particle, \mathbf{v}_p is velocity of particle, \mathbf{u}_a is velocity of gas, A_d is the profile area of particle, C_D is drag coefficient and computed from the following correlation:

$$C_D = 24(1 + 0.15 Re^{0.687}) / Re \quad (26)$$

Re is the particle Reynolds number and defined in Eqn. (27):

$$Re = \frac{\rho_g |\mathbf{u}_a - \mathbf{v}_p| d_p}{\mu} \quad (27)$$

particle energy equation:

$$m_p c_{p,p} \frac{dT_p}{dt} = H_c + H_r + H_s \quad (28)$$

where $c_{p,p}$ is specific heat capacity of the particle, T_p is temperature of particle, H_c is convective heat source for a single particle, H_r radiative source for a single particle, H_s chemical reaction heat source.

H_c is given by

$$H_c = Nu \cdot \pi \lambda_p (T_g - T_p) \quad (29)$$

where λ is the gas thermal conductivity, T_g is the temperature of gas.

Nu is the Nusselt number which can be expressed as

$$Nu = 2.0 + 0.6 Re_g^{0.5} \left(\mu \frac{c_p}{\lambda} \right)^{1/3} \quad (30)$$

where Re_g is Reynolds number of gas, c_p is the specific heat capacity of gas.

H_r is given by

$$H_r = \varepsilon_p \pi d_p^2 \left[\frac{G}{4} - \sigma T_p^4 \right] \quad (31)$$

where ε_p is particle emissivity, G is irradiance, σ is Stefan-Boltzmann constant.

H_s is given by

$$H_s = \Delta H_i \frac{dm_i}{dt} \quad (32)$$

where ΔH_i is total heat of reaction per unit mass of reactions, m_i is mass of different particle composition.

5 OVERALL SOLUTION ALGORITHM

Fig. 2 shows the overall PSIC strategy to solve the gas and particle equations. All input data were read including details of gas and particle compositions, particle sizes, boundary conditions and

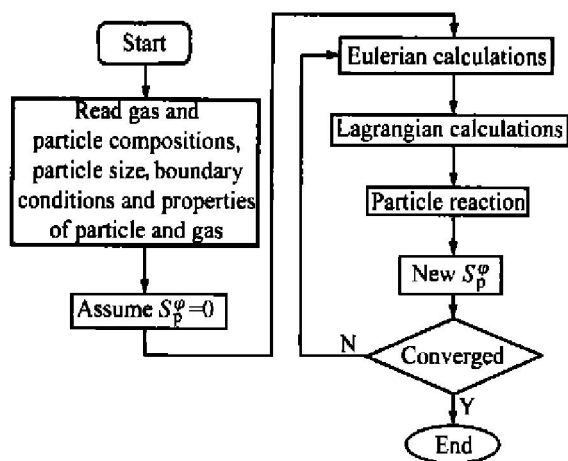


Fig. 2 Overall solution algorithm of mathematical model

properties of particle and gas. A first assumption of the source terms in the gas phase equations to zero is made, and gas equations are solved in Eulerian framework under this assumption. When the Eulerian equations are converged, the particle equations are calculated in Lagrangian framework and the reactions of particle are calculated. New source terms for all gas phase equations are computed and these source terms are under-relaxed before returning to the Eulerian calculations. One cycle of the algorithm in which the Eulerian calculations are converged and the Lagrangian calculations are computed in the new source terms is called a particle iteration. Overall convergence is achieved when the source terms do not change from one particle iteration to the next.

6 VERIFICATION OF MODEL BY PLANT TEST

It's essential to make assumptions and simplifications in modelling. However, some assumptions and simplifications will cause the great error between the fact and calculated result. Plant test is the best method to verify the model. The most important factor is temperature in flash smelting furnace, so comparison between the calculation temperature and real temperature in flash smelting furnace can verify the accuracy of model. Table 2 and Fig. 3 show the comparison between plant test temperature and calculation temperature in Guixi Flash Smelter and Jinlong Flash Smelter.

Table 2 Comparison between plant test temperature and calculation temperature in Guixi Flash Smelter

Distance from reaction shaft top/m	Test temperature/ K	Calculation temperature/ K
1.5	1 546	1 571
2.0	1 540	1 568

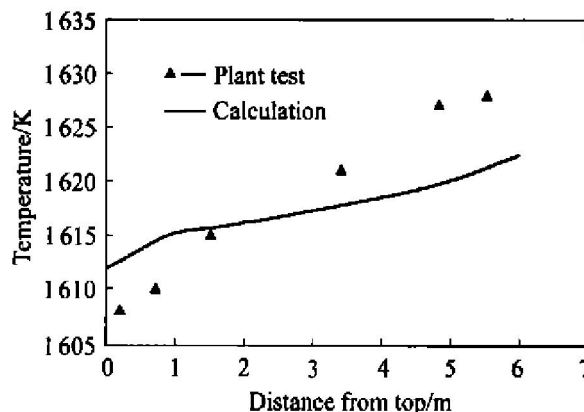


Fig. 3 Comparison between plant test temperature and calculation temperature in Jinlong Flash Smelter

Table 2 shows that the error between temperature of plant test in Guixi Flash Smelter and calculation temperature is less than 30 K. Fig. 3 shows that the error between temperature of plant test in Jinlong Flash Smelter and calculation temperature is less than 10 K. It is concluded that the mathematical model is reasonable and is able to be used to study the flash smelting process. This model is also able to be used to optimize operation in flash smelting furnace.

7 CONCLUSIONS

- 1) A description of multi-stage and multi-phase chemical reaction in flash smelting furnace is set up, and this description is more better than the fore assumption of one-step reaction.
- 2) A fully three-dimensional, turbulent and multi-phase model using momentum, mass, heat transfer and chemical reactions of the copper flash smelting furnace as parameters has been developed. This model is accurate and can be used to direct and optimize the production of flash smelting furnace.

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