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MATHEMATICAL MODELING AND SIMULATION OF DIRECT REDUCTION OF IRON ORE IN A MOVING BED REACTOR BY THE SINGLE PARTICLE MODEL

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Abstract. In this work, a mathematical model is developed for simulating the behavior of a counter-current moving bed reactor, in which the reduction of porous iron ore pellets to sponge iron is simulated. Simultaneous mass and energy balances within both the solid particles and the reactor, will lead to a set of coupled ordinary differential equations. The iron ore reduction kinetics was modeled with a single particle model. The model was able to satisfactorily reproduce the data of Gilmore Steel Corporation (USA). Eventually, the effects of reducing gas parameters and pellet characteristics such as porosity on the reduction extent have been investigated.

Keywords: simulation, direct reduction, moving bed reactor, single particle model, porosity.

1. Introduction

The direct reduction is a significant process to produce metallic iron from iron ore using reducing gases. In the moving bed solid-gas reactor the gas flow upward and the solids flow downward by gravity in a counter current. Mass and heat transfer between the phases, as well as chemical reaction occur simultaneously, which reduce the hematite pellets. In the last few years, the direct reduction of iron ore process has been investigated by using some mathematical models.

Szekely and El-Tawil [1], Towhidi and Szekely [2], Parisi and Laborde [3] used the unreacted shrinking core model to model hematite pellets. This model was in accordance with the experimental data, although it should be noted that the variation of solid porosity as a main parameter is not considered in the model. Recently, Arabi and Hashemipour [4] modeled the moving bed direct reduction reactor by the three shrinking core model, Nouri *et al.* [5] modeled it by the grain model for industrial plants. Both models satisfactorily fitted with the experimental data, but it should be noted these models do not consider the variation of solid porosity. Melchiori and Cunu [6], Ahn and Choi [7] carried out the numerical comparison between the shrinking core model and the grain model. They found that a grain model has a better performance than a shrinking core model to predict experimental and industrial results.

Moreover, some researchers assumed the direct reduction reactor model including only one reacting gas. Most of them have used pure hydrogen [8, 9], pure carbon monoxide [10] or a mixture of hydrogen and carbon monoxide as a reducing gas [11, 12], but in fact the reducing gas is a mixture of hydrogen, carbon monoxide, carbon dioxide, steam and methane as a synthesis gas.

In this work, the moving bed direct reduction reactor has been modeled by a single particle concept. This model considers the variation of solid porosity. The model equations were derived from mass and heat balance of the solid particle and bed of the reactor. The modeling results have been compared with Gilmore planet data [3] to indicate the model validation. This simulation is used to study the effect of operating parameters on the reactor performance.

2. Mathematical Model

2.1. Chemical Reaction

The reaction of hematite to iron is carried out through the series of reactions: hematite → magnetite → wustite → iron. The overall reaction which is considered in the several simulation works [3-5] is a direct reduction of hematite to iron with the reducing gases hydrogen and carbon monoxide as R1 and R2:

$$\text{R1: } Fe_2O_3 + 3H_2 \rightarrow 2Fe + 3H_2O \qquad (1)$$

$$\text{R2: } Fe_2O_3 + 3CO \rightarrow 2Fe + 3CO_2 \qquad (2)$$

The reaction of hematite with other reducing gases such as CO_2, H_2O and CH_4 is not considered in this study because of low content and negligible reactivity. The

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chemical reaction rates of the reactions R1 and R2 are introduced as elementary with the following reaction rate equations [3]:

$$R_{R1} = k_{H_2} C'_{H_2},$$

$$k_{H_2} = 0.114\exp(-179.13/T),\ \text{cm/s} \qquad (3)$$

$$R_{R2} = k_{CO} C'_{CO},$$

$$k_{CO} = 0.283\exp(-342.43/T),\ \text{cm/s} \qquad (4)$$

2.2. Particle Modeling

In this work, a modified volume reaction model named a single particle model is used as a new approach for consideration of pores structural changes within the solid phase. Molar volumes of hematite and iron are different and this causes structural changes of the solid during the reaction. In this model, the reaction of spherical solid particle with the reactant gases is considered isothermal. In addition, it is assumed that the particle diameter remains constant during the reactions. The mole balances of H_2 and CO components within the solid particle base on the model are as follows:

$$\frac{1}{r^2}\cdot\frac{\partial}{\partial r}\left(r^2 D_{eH_2}\frac{\partial C'_{H_2}}{\partial r}\right) - (R_{R1}S_g) = 0 \qquad (5)$$

$$\frac{1}{r^2}\cdot\frac{\partial}{\partial r}\left(r^2 D_{eCO}\frac{\partial C'_{CO}}{\partial r}\right) - (R_{R2}S_g) = 0 \qquad (6)$$

The boundary conditions for the above equations are as follows:

$$\text{At } r = 0 \quad \frac{dC'_{H_2}}{dr} = \frac{dC'_{CO}}{dr} = 0 \qquad (7)$$

$$\text{At } r = R_p \quad C'_{H_2} = C_{H_2},\ \ C'_{CO} = C_{CO} \qquad (8)$$

where C_{H2} and C_{CO} are the reactant gases concentration in the bulk phase (in the reactor). Mole balance on the hematite within the solid particle gives the following equation:

$$\frac{dC_C}{dt} = R_C = 3R_{R1} + 3R_{R2} \qquad (9)$$

where C_C is the hematite concentration in the particle which is $C_C = \frac{V_p e_p r_p}{M_w}$.

The Eq. (9) can be rearranged as:

$$\frac{de_p}{dt} = \frac{3M_w}{V_p r_p}(R_{R1} + R_{R2}) \qquad (10)$$

With the initial condition $e_p = e_p^0$ in Eq. (10), the time variable is defined as a distance of solid travel from its inlet (t = Z/u_s). Therefore the initial condition of solid porosity is the inlet solid porosity of the reactor.

In the single particle model, the solid particle properties (e_p, r_p and S_g) are variable because of structural changes within the particle [13]. In this simulation, a second order functionality of a surface area versus particle porosity is considered. This relation is derived using the mathematical points (e_p = 0, S_g = 0), (e_p = 1, S_g = 0) and ($e_p = e_p^0$, $S_g = S_g^0$) as the following equation:

$$S_g = S_g^0\frac{(e_p - e_p^{\,2})}{(e_p^0 - e_p^{02})} \qquad (11)$$

The particle density is also variable as the reaction progresses and the particle porosity change. This functionality is defined as $r_p = r_t(1–e_p)$. The value of r_t is a true density of the solid. The pore diffusion of gas component within the solid particle (D_{ei}) is a function of the particle porosity. This relation can be evaluated using expression [14]:

$$D_{ei} = D_i e_p^{\,2} \qquad (12)$$

The gas diffusion coefficients as a function of the solid temperature can be evaluated using expressions [3]:

$D_{H2} = 1.467\cdot 10^{-6}\cdot T_s^{1.75}$, cm²/s (13)

$D_{CO} = 1.276\cdot 10^{-7}\cdot T_s^{1.75}$, cm²/s (14)

The above mathematical modeling of the particle leads to a set of coupled nonlinear ordinary differential equations.

2.3. Reactor Modeling

In this study a non-catalytic moving bed reactor is modeled and simulated in the non-isothermal condition. In the moving bed reactor, the gas flows upward and countercurrent to the downward flow of solids. Some of the general assumptions such as steady state operation and plug flow of the gas and solid streams in the reactor are considered to derive the reactor models. The mass balance of gaseous reactants within the reactor can be written as follows:

$$u_g\frac{dC_{H_2}}{dz} + \frac{1-e_b}{V_p}J_{H_2} = 0 \qquad (15)$$

$$u_g\frac{dC_{CO}}{dz} + \frac{1-e_b}{V_p}J_{CO} = 0 \qquad (16)$$

The mass balance on the reactant solid can be written as follows:

$$u_s\frac{dC_C}{dz} + \frac{1-e_b}{3V_p}(J_{H_2} + J_{CO}) = 0 \qquad (17)$$

J_{H2} and J_{CO} are described as diffusion rates of the gas components into the solid particle. These factors can be evaluated using expressions:

$$J_{H_2} = -D_{eH_2} e_p A_p \frac{dC'_{H_2}}{dr}(r = r_p) \qquad (18)$$

$$J_{CO} = -D_{eCO} e_p A_p \frac{dC'_{CO}}{dr}(r = r_p) \qquad (19)$$

The gas component concentration within the solid particle is predicted using the single particle model. The energy balance for gas and solid phases can be stated as follows:

$$G_{mg}C_{pg}\frac{dT_g}{dz}+\frac{A_p}{V_p}(1-e_b)h(T_g-T_s)=0 \qquad (20)$$

$$G_{ms}C_{ps}\frac{dT_s}{dz}+\frac{(1-e_b)}{V_p}[A_p h(T_g-T_s)+$$

$$+\Delta H_{H_2}J_{H_2}+\Delta H_{CO}J_{CO}=0 \qquad (21)$$

Boundary conditions for the above equations are as follows:

$$\text{At } z = 0 \quad C_{H_2}=C_{H_2}^{in},\ C_{CO}=C_{CO}^{in},\ C_C=C_C^{in} \qquad (22)$$

$$\text{At } z = 0 \quad T_g=T_g^{in},\ T_s=T_s^{in} \qquad (23)$$

The heat capacity of the gas and solid components are considered variable as the temperature variation. The heat transfer coefficient and heat capacity of the gas and solid components used in the simulation are shown in Table 1.

Table 1

The heat transfer coefficient and heat capacity of the components

Symbols, units	Values
$C_{p_{Fe2O3}}$, cal/mol·K	24.72+0.01604T–423400/T^2
$C_{p_{H2}}$, cal/mol·K	6.62+0.00081T
$C_{p_{CO}}$, cal/mol·K	5.12+0.00333T
h, cal/cm^2·s·K	1·10^{-4}

The solid conversion can be evaluated using the expression:

$$X=1-\frac{C_C}{C_C^{in}} \qquad (24)$$

2.4. The Numerical Solution

The nonlinear ordinary differential equations of the moving bed reactor and solid particle are solved simultaneously. These equations describe variation of the gas component in the reactor and solid particle. The gas concentrations in the reactor are related to the gas concentrations profile within the solid particle with the pore changes. These nonlinear differential equations are solved using numerical algorithm programmed with Matlab software. The software code is programmed using ODE solution algorithm.

3. Results and Discussion

3.1. Simulation Validation

In this section, a comparison between the results of the simulation model with the values of existing gas composition and metallization of an industrial plant is performed. The model is validated using experimental data from Gilmore plant [3]. The operating conditions of the plant reactor and the inlet gas composition are presented in Table 2. The reactor simulation was run with these parameter and the results are shown in Fig. 1.

The outlet gas composition (Fig. 1, length 0 cm) and outlet solid conversion (Fig. 1, length 975 cm) predicted from the simulation are compared with the experimental data. The comparison is presented in Table 3. Results of the simulation model are in good agreement with the plant data with an average error of 3.2 %.

In the next sections, the effects of some operating conditions of inlet streams on the solid conversion are investigated. To study the effect of these parameters, the amounts are selected based on the Gilmore operating conditions.

3.2. Temperature Profiles of Gas and Solid within the Reactor

Basical temperature profile along the reactor (especially solid temperature) is the main parameter in the reactor operation because it affects the reduction progress directly. Fig. 2 illustrates the temperature profiles of gas and solid phases along the reactor. The temperature of gas stream decreases from bottom (gas inlet) toward the top of the reactor, while the temperature profile of solid stream has an ascending trend with a higher slope. This is because of heat transfer from the gas to the solid in addition to the endothermic reactions within the solid particles.

The effects of temperature of the gas and solid inlet on the solid conversion along the reactor are shown in Figs. 3 and 4. Fig. 3 illustrates that increasing gas inlet temperature improves the solid conversion but there is a negligible effect of the higher gas inlet temperature. This can be for higher diffusion limitation at the higher temperature. Therefore the optimum temperature of the gas inlet is about 1000 K. Fig. 4 illustrates that increasing solid inlet temperature has less effect on the solid conversion improvement (in the temperature range of 325–525 K). The reason is that this temperature range is too low to affect the chemical reaction.

Table 2

Operating conditions of Gilmore plant [3]

Parameters	Values
Gas flow rate, Nm^3/h	53863
Inlet gas composition, wt % (at $z = L$)	
H_2	52.58
CO	29.97
H_2O	4.65
CO_2	4.80
CH_4+N_2	8.10
Pressure, kPa	141.8
Gas inlet temperature, K	1230
Solid flow rate (Fe), t/h	26.40
Iron ore particle density (r_t), g/cm^3	4.70
Sponge iron particle density (r_p), g/cm^3	3.20
Particle radius (R_p), cm	0.55
Inlet particle porosity (e_p)	0.30
Solid inlet temperature, K	320
Reaction zone length (L), cm	975
Reactor diameter (R), cm	426
Bed porosity (e_b)	0.55

Table 3

Comparison of Gilmore data with the developed model prediction

	Gilmore data, wt %	Developed model data, wt %	Error, %
Outlet gas composition H_2	37.0	37.2	0.53
CO	18.9	18.7	1.06
H_2O	21.2	20.5	3.41
CO_2	14.3	15.5	7.74
Inert	8.6	8.1	6.17
Solid conversion	93.0	92.7	0.32

3.3. Variation of Gas and Solid Flow Rates

Fig. 5 illustrates the effect of inlet reducing gas flow rate on the conversion of iron ore particles in the range of ±10 %. When the gaseous flow rate is decreased, a lower solid conversion is obtained. This is due to decreasing of reducing gaseous content in the reactor as the gas flow rate is decreased. In this condition, the overall reaction rate decreases and therefore the solid conversion gets worse.

Fig. 6 illustrates the effect of variation of the inlet solid flow rate in the range of ±15 % on the conversion of iron ore particle. As the solid flow rate is decreased, the conversion of outlet particles is increased because the residence time of iron ore particles in the reactor is increased. Since the reaction is happened in the solid phase, so the solid particles have more time to react with the gaseous reactants.

In other words, it can predict that in this system the gas content should be an excess and in this condition the effect of this parameter on the solid conversion is low, while the solid is consumed along the reactor considerably and so a solid flow rate is more important than a gas flow rate on the reactor performance.

3.4. Feed Gas Composition

Generally the inlet gas stream contains H_2, CO, CO_2, H_2O, CH_4 and N_2 which is a product of the syngas production unit where H_2 and CO components are more effective than the others in the reduction of hematite. In this study these two components are considered to reduce the solid. Therefore the feed gas composition named as a molar ratio of H_2/CO is a main parameter in the prediction of solid reduction but it is highly restricted. This is due to decreasing the low mass transfer of the reducing gas into the solid.

Fig. 7 illustrates the effect of this molar ratio on the solid conversion. The solid conversion is decreased with the decrease in CO concentration (with increasing H_2/CO ratio) in the feed gas. Since the rate constant for CO is higher than that of hydrogen at the same temperature, therefore the higher CO content is preferred from the reaction viewpoint. But in the real condition, removing of H_2 from the gas inlet causes the coke formation on the sponge iron due to decomposition of CO component and therefore this parameter cannot vary in a wide range.

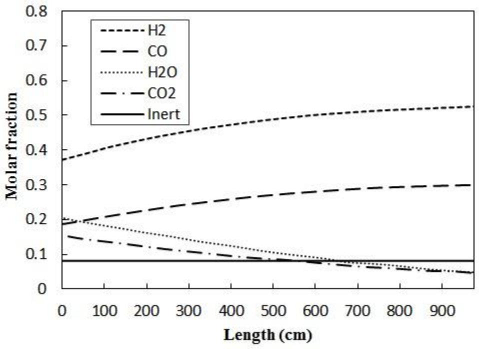

Fig. 1. Profile of gas composition along the reactor

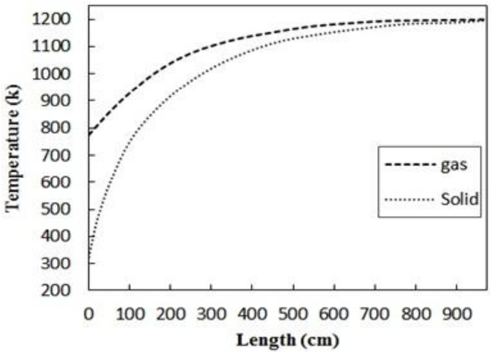

Fig. 2. Variation of solid and gas temperature along the reactor

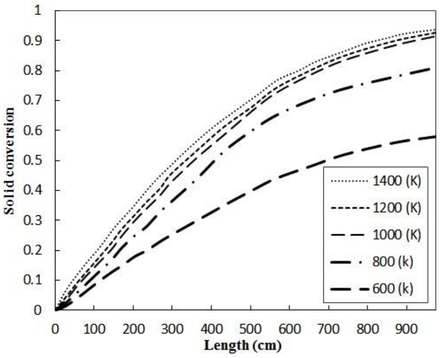

Fig. 3. Effect of gas inlet temperature
on the conversion of iron ore

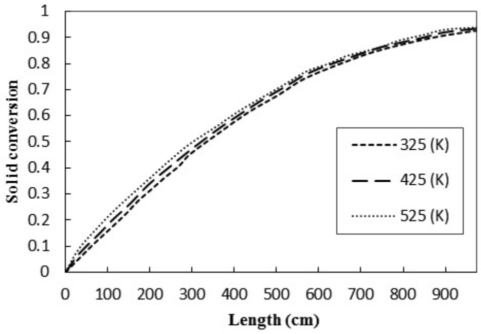

Fig. 4. Effect of solid inlet temperature
on the conversion of iron ore

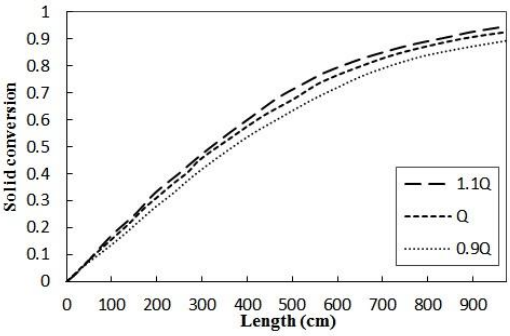

Fig. 5. Effect of gas flow rate on the conversion
of iron ore (Q = 53863 Nm^3/h)

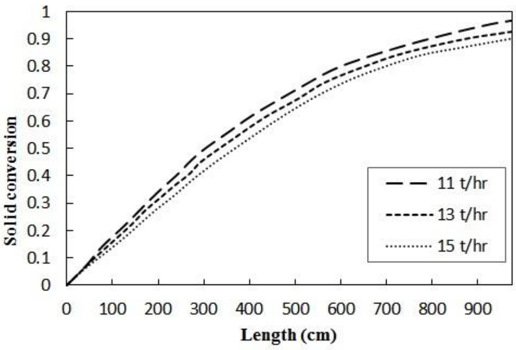

Fig. 6. Effect of solid flow rate
on the conversion of iron ore

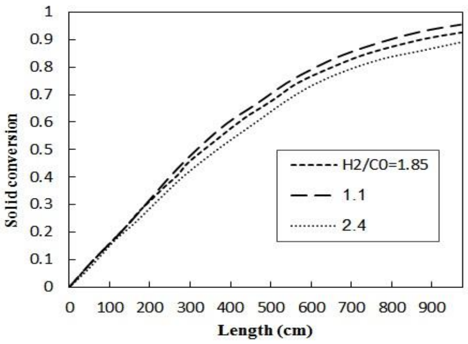

Fig. 7. Effect of feed gas composition
on the conversion of iron ore

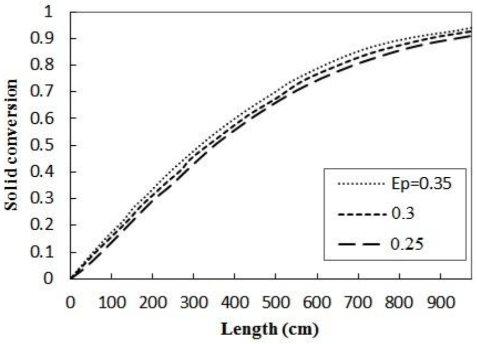

Fig. 8. Effect of particle porosity
on the solid conversion

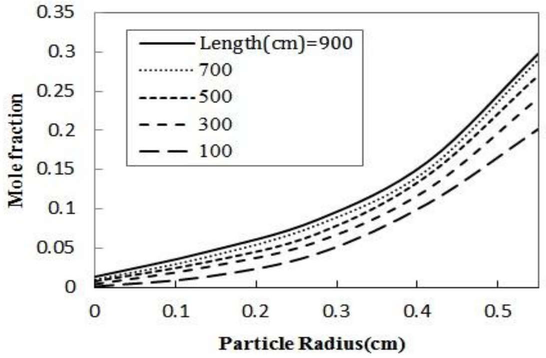

Fig. 9. Gas composition (CO) profile inside the particle in some different reactor length

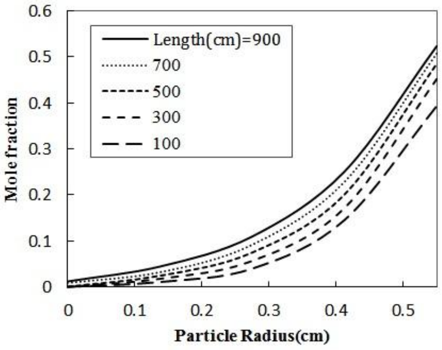

Fig. 10. Gas composition (H_2) profile inside the particle in some different reactor length

3.5. Effect of Inlet Particle Porosity

The particle porosity increases as the reduction progress due to the difference in molar volume of hematite (as reactant) and iron (as a product). This variation of porosity is very important because the reaction surface and pore diffusivity, as well, are related to porosity directly. The initial porosity should be in the pre-determined amount and it can not vary in the wide range. The effect of inlet solid porosity on the conversion of solid is shown in Fig. 8. It can be seen that when the inlet particle porosity is increased, an increasing in solid conversion is observed. This is because of higher surface area and lower gas diffusion resistance in the higher porosity of solid. This condition causes improvement of overall reaction rate.

3.6. Gas Composition Profile inside the Particle

Figs. 9 and illustrate gases compositions (CO and H_2) profile within the solid particle in several points in the reactor length. It can be seen that the slope of composition profile for both gases is decreased from an external surface to the center of the particle. These slopes are related strongly to the gases compositions in the bulk flow within the reactor. In addition, solid porosity affects the gas diffusivity and thus the gas concentration profile within the solid particle. These factors explain the effect of a reactor length on the gas concentration profile within the solid particle. Because of counter current flow of gas and solid stream in the reactor, the effect of length on the gas profile within the particle is not vigorous. This weak effect is observed in both figures.

4. Conclusions

In this work, reduction of iron ores in the moving bed reactor was simulated by the single particle model. In order to simulate the system, the equations of mass and energy balances into the reactor and within the particle are derived for the solid and gases components. Then the equations were solved in the countercurrent gas-solid flow. The results of the simulation were in a good agreement with the experimental data of Gilmore plant data (average error 3.2 %). The effects of reactor and solid particle parameters were investigated by the simulation. The results of the simulation show that the inlet reducing gas parameters (composition, temperature and flow rate) and the porosity of the inlet solid are the main and effective parameters on the solid conversion. In addition, the optimum amount of the reactor length was 900 cm and the optimum gas inlet temperature was 1000 K. Comparison of the gas concentration profile within the solid particles in different reactor length showed that the gas diffusion within the solid particles had the main effect on the reaction progress.

Nomenclature

A_p	outer surface area of the particle	L	total reactor length	X	solid conversion	C	molar concentration within the gas phase
C_{pg}, C_{ps}	heat capacity for gas and solid phases	n_p	number of particle per unit volume of the bed	ΔH	heat of reaction	C^{in}	initial molar concentration
D	molecular diffusivity	Q	gas flow-rate	e_p	particle porosity	S_g	specific surface
D_e	effective diffusivity of gas	r	particle radius	e_p^0	initial particle porosity	S_g^0	initial specific surface
G_{mg}, G_{ms}	gas and solid molar flows	T_g, T_s	gas and solid phase temperatures	e_b	reactor bed porosity	r_p	initial particle density

h	heat transfer coefficient	t	time	Z	reactor length	r_p	total particle radius
k	reaction rate constant	u_g, u_s	gas and solid velocity	C'	molar concentration within the solid particle	M_w	molecular weight
						V_p	particle volume

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МАТЕМАТИЧНЕ МОДЕЛЮВАННЯ ТА СИМУЛЯЦІЯ ПРЯМОГО ВІДНОВЛЕННЯ ЗАЛІЗНОЇ РУДИ В РЕАКТОРІ З РУХОМИМ ШАРІ НА МОДЕЛІ ОКРЕМОЇ ЧАСТИНКИ

Анотація. *Розроблено математичну модель для симуляції відновлення пористих залізорудних окатишів до губчастого заліза в протиточному реакторі з рухомим шаром. На основі масового та теплового балансів як в твердих частинках, так і реакторах, виведено звичайні диференціальні рівняння. За моделлю окремої частинки змодельована кінетика відновлення залізної руди. Показано, що розроблена модель задовільно відтворює дані корпорації* Gilmore Steel *(США). Досліджено вплив параметрів відновлювального газу та характеристик окатишів, таких як пористість, на ступінь відновлення.*

Ключові слова: *симуляція, пряме відновлення, реактор з рухомим шаром, модель окремої частинки, пористість.*