ANALYSIS OF CHEMICAL-METALLURGICAL AGGLOMERATION PROCESSES DURING CHARGE SINTERING

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ABSTRACT

Thermally activated chemical and metallurgical processes of lumping of fine-dispersed iron ore raw material during sintering are examined in the work. The following processes are included: moisture removal, ignition of coke fines and burn-off of carbonates, taking into account burning features of coke fines, fusion of grain charge, forming of agglomeration cake, creation of overhumidification area in sintering layer owing to condensation of water vapours in washout of heat-carrying gas. Parameters of sintering kinetics are obtained experimentally, other parameters characterizing layer shrinkage of sintering charge are presented. It was established that the temperature in the sintering area has substantial effect on shrinkage. The presented temperature relationship describes experimental data with sufficient precision. The values of power exchange coefficients in the drying area are examined. The criteria equations of heat and mass exchange are presented.

The mathematical model using empiric material is proposed; it describes sintering kinetics for agglomeration charge, taking into account layer shrinkage and variation of head losses in technological areas of fusion and forming of ready agglomerate, what corresponds well with the experimental data. Multi-factor relationship between chemical and metallurgical processes (from one side) and heat and mass exchange in the sintering layer of agglomeration charge (from other side) is noted. Adequacy of mathematical models allows to use them for analysis of high-efficient conditions of agglomerate fabrication.

Introduction

Agglomeration of fine-dispersed raw materials at belt-type conveyors is the main method of lumping of ore fines for metal production in metallurgical industry. Fine-dispersed ore raw materials is mixed with coke fines and corresponding components necessary for obtaining of the required properties of finished product; then it is preliminarily pelletized and laid in the sintering layer of charge at the conveyor of a sintering machine (presented by special sintering cars - pallets) [1]. Then the upper level of charge is heated up to the temperature of fuel ignition while moving in the hearth of a sintering machine. After processing in the hearth, the sintering charge is subjected to air washout, what provides steady burning [2]. Speed of conveyor motion is determined by the conditions of burning finishing [3]. Heated agglomeration cake is transferred to crusher and sieve and afterwards the obtained lump material is subjected to cooling [4].

As soon as agglomeration in metallurgy is accompanied by essential consumption of raw materials and power, the problem of power supply is rather actual.

The aim of this work is the complex analysis of mutually depending chemical and metallurgical processes occurring during agglomeration of iron ore fines, in order to improve power and resource efficiency.

Conditions of analyzed agglomeration process

Analysis of agglomeration process was conducted in the following conditions: part of concentrate 61−66%; part of carbon in charge 4%; charge humidity 5−7%; return part 25−30%; height of charge layer 150−200 mm.

Material and heat balance of the process of iron ore fines agglomeration (per 100 kg of agglomerate) are presented in the **tables 1 and 2**.

Attention should be attracted in consumption items to heat of waste gases, heat of agglomeration cake and wastes in the environment.

Physical and chemical transformations in raw materials during agglomeration

Agglomeration of fine-dispersed iron ore materials is a high power-efficient technological process meaning its heat and power conditions [5]. The sintered layer in the burning area is practically completely melted at rather

Table 1. **Material balance of agglomeration process**					
Income item	kg	%	Consumption item	kg	%
Iron ore materials and metal additived	74.94	25.56	Agglomerate	100	34.1
Flux	31.32	10.68	Waste gases	190.04	64.81
Coke fines	11.65	3.97	Agglomeration dust (slime)	3.2	1.09
Gas for ignition	0.88	0.3			
Air for sintering	157.06	53.56			
Charge moisture	17.39	5.93			
Total amount	293.24	100	Total amount	293.24	100

Table 2. Heat balance of agglomeration process

Income item	кДж	%	Consumption item	кДж	%
Fuel burning	236014	82.55	Evaporation of hygroscopic moisture	39258	13.73
Fe_2S and FeS	1828	0.64	Removal of hydrate moisture	11788	4.12
Charge heat	2963	1.04	Dissociation of carbonates	19060	6.67
Air heat	3133	1.10	Decomposition of complicated minerals	983	0.34
Ignition	34000	11.89	Heat of waste gases	55052	19.26
FeO oxidation	919	0.32	Heat of cake	101174	35.39
Silicate forming	7050	2.47	Charge melting	15493	5.42
			Heat losses	43099	15.07
Total amount	285907	100	Total amount	285907	100

small consumption of coke fines (4−8%) and is consequently crystallized as porous cake (agglomerate) [6].

Heat power can be regenerated based on rational organization of the technological process, what allows to rise power efficiency approximately by 40%. Removal of moisture is rather power-intensive process, and it has significant effect on power engineering and technology of agglomerate sintering in the layer [7].

Mutual influencing physical and chemical transformation processesб being essentially depended on the conditions of heat and mass exchange [8] are occurred simultaneously at different heights of sintered layer in agglomeration charge. The following transformations can be mentioned here: moisture evaporation, ignition of coke fines, burning of coke fuel and fusion of charge particles, endothermic decarbonization and other chemical reactions, condensation of moisture vapours in the lower levels of sintered layer, agglomeration cake forming.

High intensity of heat exchange between heat-carrying gas and charge particles can be provided owing to rather developed surface of charge particles and fuel burning inside sintered layer; high power efficiency of heat power usage is achieved as well [9]. In agglomeration process, air for fuel burning is washing out through sintered layer and provides heat recuperation from more heated upper levels if sintered layer of agglomeration charge to less heated lower levels. Therefore, heat and power exchange can be characterized as regeneration process [10]. Speed of transition of agglomerate in burning area from top downward in the sintered layer depends on heat and mass exchange as well as physical and chemical transformations and is described by the heat balance equation for elementary layer.

$$\Delta h \rho_m (1-\varepsilon) c_m \Delta t_m = w_g \Delta \tau \rho_g c_g \Delta t_g ,$$

where Δt_m, Δt_g — temperature variations of charge materials and heat-carrying gas on the elementary layer section with height Δh, during elementary time period $\Delta \tau$; c_g, c_m — gas heat capacity and apparent charge heat capacity, including heat absorption and extraction as a result of en-

dothermic and exothermic processes; ε — porosity of sintered layer; w_g — washout speed of heat-carrying gas; ρ_m — density of charge material; ρ_g — density of heat-carrying gas. Dimensions of all values included in the equations are suggested to be in the International System of Units (SI).

Sintering speed is proportional to filtration speed of heat0carrying gas and depends on charge material properties

$$u = \Delta h / \Delta \tau = \left(w_g \rho_g c_g \Delta t_g \right) / \left(\rho_m (1-\varepsilon) c_m \Delta t_m \right) .$$

In general case, it is necessary to evaluate heat capacity of charge material c_m for assessment of sintering and agglomerate fabrication speed, taking into account kinetic features of phase transformations in charge, fuel burning and non-isothermal processes of carbonates dissociation with heat effects of decomposition of $CaCO_3$ ($Q = 1778$ kJ/kg) и $MgCO_3$ ($Q = 1395$ kJ/kg). Kinetics of decarbonization is considered by the authors in [11, 12] and obtained results were used for analysis of dissociation processes of carbonates during agglomeration.

Fuel burning in sintered layer of agglomeration charge has its own features. This process is usually presented as two reactions occurring simultaneously:

a) $C + O_2 = CO_2 + 410$ MJ/kmol;

b) $C + 0,5O_2 = CO + 124$ MJ/kmol.

With temperature rising, the second reaction is most possible. The conditions for sold phase reduction appear. The opposite tendency can be observed at increase of gas phase pressure. The speed of oxidant feed to the surface of reacting particles has the effect on reaction process. The picture of heterogeneous burning varies depending on the relationship between oxygen diffusion processes and reaction speed.

Internal porous reaction is also possible in kinetic conditions, while only surfacial reaction can be observed in diffusion conditions. The constant of specific surfacial speed of carbon burning can be presented as $k_s^c = \beta_c C_{O_2} (1/k + 1/\alpha_D)^{-1}$ where: β_c — stoichiometric coefficient within the range 0.37−0.75 depending on reaction via the mechanism a) or b); C_{O_2} — oxygen concentration; k — the constant of burning speed of a solid fuel particle; α_D — mass exchange coefficient determining via criteria equations: for freely burning particles $Nu_D = 2(1 + 0,08Re^{2/3})$. For the particles burning in layer: $Nu_D = 0,187Re$, when $7 \le Re \le 27$ or $Nu_D = 0,01Re^{2,5}$, when $Re < 7$; where $Nu_D = \alpha_D d_c / D$, d_c — coke particle diameter; D — oxygen diffusion coefficient.

The equation describing burn-off of a coke particle in layer is as follows:

$$d \left(\rho_c \pi d_c^3 / 6 \right) = -k_s^c \pi d_c^2 d\tau .$$

The main task of agglomeration technology of iron ore fines is lumping. Agglomerate lumps should meet the requirements in strength, they also should be characterized by easy reducibility. Agglomerate forming via sintering occurs in the temperature range

Table 3. Values of parameters for shrinkage evaluation of sintered charge layer

Size of particles, mm	t, °C	$\mu_m \cdot 10^{-6}$, Pa·s	n	Size of particles, mm	t, °C	$\mu_m \cdot 10^{-6}$, Pa·s	n
0.14–0	1250	4.4	0.68	0.2–0.14	1100	230	0.78
0.2–0.14	1250	4.4	0.68	0.063–0	1200	20	0.72
0.14–0	1200	12	0.74	0.1–0.063	1200	20	0.72
0.2–0	1200	12	0.74	0.16–0.1	1200	20	0.72
0.2–0.14	1200	12	0.74	0.063–0	1150	42	0.75
0.14–0	1150	38	0.76	0.1–0.063	1150	42	0.75
0.2–0	1150	38	0.76	0.16–0.1	1150	42	0.75
0.2–0.14	1150	38	0.76	0.063–0	1100	48	0.76
0.14–0	1100	230	0.78	0.1–0.063	1100	48	0.76
0.2–0	1100	230	0.78	0.16–0.1	1100	48	0.76

800−1500 °C. Sintering of agglomeration charge starts from solid phase processes and finishes by melt forming. Development of solid phase processes depends on properties of iron ore raw material and thermal conditions of lumping, while thermal conditions provide charge melting; there is no other way to provide fabrication of strong agglomerate.

The main regularities of the sintering theory are described in the works of different authors [13−15] and, according to them, sintering can be realized via the following ways [16]:

tough flow, i.e. via directed transition of substance to a contact isthmus with increase of contact square;

volumetric and boundary diffusion, i.e. via transition of substance with increased contact of particles without approximation of their centers;

transition of substance through gaseous phase owing to difference between equilibrium vapours pressure near concave and convex sections of shapes of contacting particles.

Simulation of chemical and metallurgical processes occurring in charge during heating

Use of the above-mentioned mechanisms in the sintering models allow to obtain relationship $(x/r_m)^n = A\tau$, where x — radius of sintering area, τ — time; r_m — radius of sintering particles; $n = 2$, $A = 1.5\sigma_t(\mu_r r_m)$ — for the mechanism of tough flow; $n = 5$, $A = 20\pi\sigma_t / \left(kt_m r_m^3\right)$ — for volumetric self-diffusion with drain of vacancies to the surface; $n = 5$, $A = 80a^3\sigma_t D_p / \left(kt_m r_m^3\right)$ — with drain of vacancies to the contact boundary; $n = 6$, $A = 48a^3\sigma_t D_B / \left(kt_m r_m^4\right)$ — for boundary self-diffusion; $n = 7$, $A = 28a^3\sigma_t \delta D_s / \left(kt_m r_M^4\right)$ — for surfacial self-diffusion; $n = 3$, $A = (28/\pi)^{1/2}\left(m/kt_m\right)^{3/2}\delta_t P_0 / \left(\rho_m r_m^2\right)$ — for transfer of substance through gaseous phase at small gas pressures; $n = 5$, $A = \left(40a^4\sigma_t P_0\right) / \left(\pi r_m^3\left(6mkt_m\right)^{1/2} P_z\right)$ — at large pressures; D_p, D_s — coefficients of boundary and surfacial diffusion; k — Bolzmann constant; a — lattice period; m — mass of a substance molecule; δ — with of the area where surfacial or boundary diffusion takes place; P_z — excessive pressure; σ_t — coefficient of melt surface tension; μ_m — dynamic toughness.

Application of rheological models allows to connect volumetric deformation and tension via the following equation

$$d\varepsilon/d\tau = (2\sigma_t)/[4/3\mu_m(1-\varepsilon)^2 r_m] \qquad (1)$$
$$\Delta V/V = \Delta\varepsilon/(1-\varepsilon).$$

Integrating the equation (1) we get

$$\ln(\varepsilon_0/\varepsilon) = 1.5(\sigma_t/r_m\mu_m)\tau.$$

Analysis of influence of lumping conditions on agglomerate strength can be conducted, taking into account correlation between porosity and strength, which is expressed by different relationships, based on the data of different authors [17, 18]. This research use the following formula in calculating experiments: $\sigma = \sigma_k[1 - 1.5\varepsilon(1 + 2k_s) + 4.5k_s\varepsilon^2]$, where k_s — empiric coefficient of attenuation; σ — agglomerate strength; σ_k — final agglomerate strength.

Discussion of experimental and numerical results obtained on the models

The results of experimental researches of sintering kinetics for iron ore fines are described well by the relationship of the following type (1):

$$\Delta l/l = 0.75(\sigma_t/r_m\mu_m)\tau^n.$$

The values of parameters for shrinkage evaluation of sintered charge are presented in the **Table 3**.

Surface tension increases and toughness decreases with rise of basicity of sintered material (acidity index was experimentally varied from 0.1 to 0.5) [19, 20]. Therefore, the tough flow mechanism prevails in sintering of agglomeration charge. Both factors provide enlargement of agglomerate strength [21].

The experimental data on kinetics of isothermal sintering (**Fig. 1**) testify that the temperature in the sintering area has essential effect on shrinkage.

The temperature relationship with sufficient practical accuracy can be described in the exponential form: $A_1 = k_s\exp(-E_c/t_m)$, where A_1 is included in the equation for shrinkage as $\Delta l/l = (A_1\tau)^n$. The values E_A and n are presented in the **Table 4**.

In the process of sintering, gas dynamic layer resistance increases, what has in its turn the effect on the conditions of mass transfer [17]. Gas dynamic resistance of sintered layer elevates by 10−15 times in comparison with initial charge layer.

Moreover, resistance values differ essentially in different layer areas [18, 22]. The following relationship is true for gas dynamic resistance:

$$\Delta P/\Delta h = \rho_{g_0} W_{g_0}(k_{g_1}\nu_{g_t} + k_{g_2} W_{g_0}[t_g/t_{g_0}]).$$

where ρ_{g_0}, W_{g_0} — density and speed of gases at normal conditions; ν_{g_t} — kinematic gas toughness at the temperature t_g; k_{g_1}, k_{g_2} — empiric coefficients.
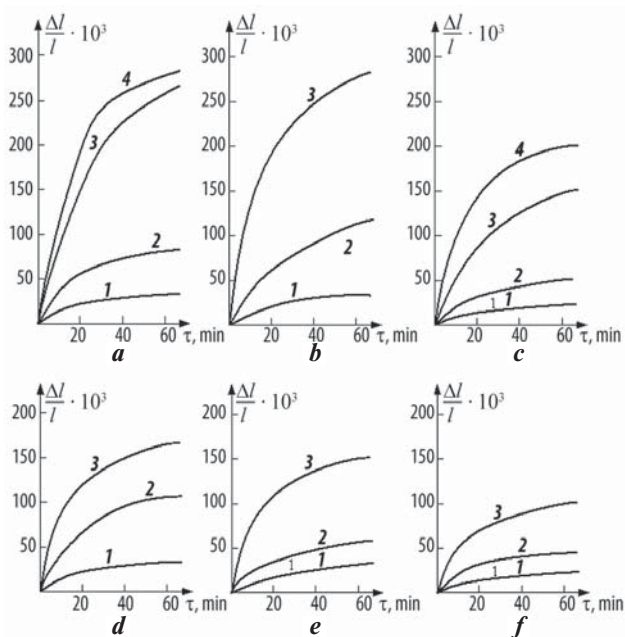
Fig. 1. Experimental shrinkage values during isothermal sintering:
1 — 1100 °C; *2* — 1150 °C; *3* — 1200 °C; *4* — 1250 °C; *5* — min; *a–c* — ore fines with lump size 0.14–0; 0.2–0; 0.2–0.14 mm respectively; *d–f* — coke fines with lump size 0.063–0; 0.1–0.63; 0.16–0.1 mm respectively

Table 4. **Values of empiric coefficients E_c and n in the equation for shrinkage depending on temperature**

	Size of particles, mm	n at the temperature, °C				$E_c \cdot 10^{-3}$, 1/K
		1100	1150	1200	1250	
Ore fines	0.14–0	0.36	0.78	0.64	0.61	39.37
	0.2–0	0.81	0.77	0.72	–	39.69
	0.2–0.14	0.79	0.80	0.70	0.60	39.5
Coke fines	0.063–0.077	0.77	0.74	0.72	–	35.8
	0.1–0.063	0.78	0.74	0.73	–	37.02
	0.16–0.1	0.79	0.76	0.68	–	38.65

Table 5. **Average values of the empiric coefficients k_{g_1}, k_{g_2} of agglomerating charge**

| Area	$k_{g_1} \cdot 10^{-5}$, m^{-2}	k_{g_2}, m^{-1}
Agglomerate cake	12–16	80–120
Melting	10–13	30–60
Intensive heating	8–10	6–20
Drying	2–5	6–14
Overhumidification	2–3	4–6
Initial charge	1–3	4–6

The average values of the coefficients for phosphoritic agglomerating charge in different areas are presented in the **Table 5**.

Temperature measurements for particles of damp agglomerate charge were conducted during its drying in four points along the layer height for examination of heat transfer coefficients. The layer height of drying charge made 130 mm. The temperature of heat carrier fed from a diesel fuel burning chamber to the layer with speed 0.3–1.0 m/s was 920 °C. Thermocouples for measuring the
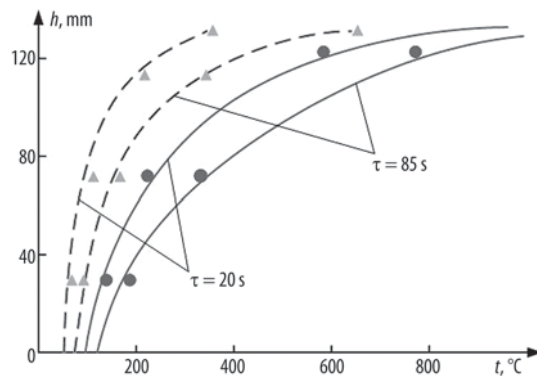
Fig 2. Distribution of gas temperature and charge during drying:
——— — gas; – – – – — charge; *1* — s
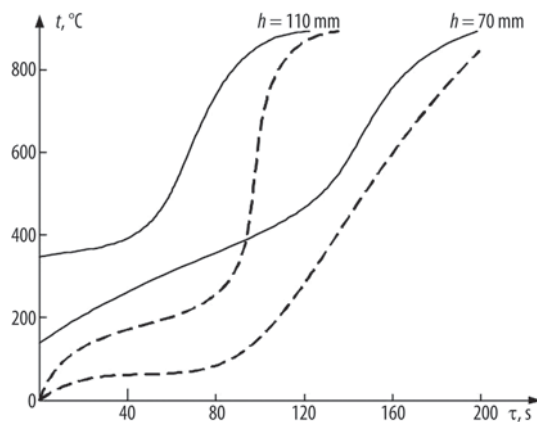
Fig 3. Values of gas temperature and charge in the sintering layer at $h = 110$ mm and $h = 70$ mm during drying process:
——— — gas; – – – – — charge; *1* — s

temperature of agglomerate charge lumps and gas were located along the layer height $h = 30, 70, 110, 130$ mm. The following graphs illustrating distribution of the temperature of heat-carrying gas and agglomerating charge along the layer height were built (**Fig. 2**).

Heat transfer coefficient α_F was evaluated from the relationship $\alpha_F = \rho_g c_g W_g \left(\partial t_g / \partial y \right) / \left(t_g - t_m \right) f_F$, where f_F — specific surface in the sintering layer.

This relationship was used at the time when the drying front reached the level, where the measurements were conducted. Evaluation of heat transfer coefficient was done at two upper levels at $h = 110$ mm and $h = 70$ mm. Location of the drying front was determined via the charge temperature field. The experimental results are presented in criteria form as follows: $\mathrm{Nu} = 2 + 0.42 \mathrm{Re}^{0.6} \mathrm{Pr}^{0.33}$. Transfer speed of the moisture evaporation front can be found through transfer of the section typical for drying finishing, when charge starts to be heated intensively (**Fig. 3**).

Comparison of charge heating conditions for two layers allowed to obtain the value of drying speed 0.81 m/s at washout speed of drying agent (heat-carrying gas) $w_g = 0.7$ m/s. Low accuracy of the results (about 30%) is explained by the errors in calculation of gas consumption at the points of thermocouples mounting, be evaluating

determination f_F of composition of burning products, by approximate building of the temperature field and by numerical differentiation.

Conclusions

It is necessary to have trustworthy information about the processes of moisture exchange and heat exchange as well as thermal hydraulics during washing out of hot gases through the layer, to provide rational holding of ignition process of coke fines [23]. Heat exchange in the dispersed layer during coke heating, coke burning and moisture removal runs very intensively owing to very developed surface in the volume of dispersed layer [24]. The processes connected with the aimed process (agglomeration cake fabrication) influence on power exchange intensity.

The mass exchange processes are determined in general by multiple factors, what pre-define building of generalized relationships, taking into account the effect of the following factors: conditions of heat exchange; layer structure; properties of substances subjected to drying; humidity of heat carrier; temperature; thermal physical properties of heat-carrying gas etc.

Obtaining of semi-empiric relationships for description of complete totality of appearances during agglomeration is directed on solving of the problems of process management and search of optimal conditions for fabrication of finished products.

Summary

Agglomeration processes allowing to evaluate adequacy of mathematical models for analysis of high-efficient conditions of agglomerate fabrication are conducted and researched.

Parameters of relationships for description of sintering and drying processes, hydraulic resistance and heat exchange coefficients are obtained.

The range of varying the initial data in the research allows to use the results for wide variety of agglomeration processes.

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