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Research

PYROMETALLURGICAL PROCESSING OF SULFIDE POLYMETALLIC RAW STUFF IN AUTOGENEOUS FORCED MODE

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ABSTRACT

An innovative approach to development of pyrometallurgical autogeneous processing of sulfide copper nickel raw stuff is proposed, which is based on radically new air blasting into melt. Spatially oriented air blasting makes it possible to combine areas of heat generation and exchange in operation space of autogeneous assembly to the greatest possible extent. In order to implement the spatially oriented air blasting a new design series of tuyeres is proposed, which provide generation of ultrasound jets outflowing at any preset angle to bath surface. Experiments with cold models and fire assemblies confirmed possibility to increase specific blasting load in two times and higher in comparison with existing methods of air blasting. The following technical effects are assumed after implementation of the scientific research results: decrease in specific power consumptions; increased time of campaign lives of autogeneous assembly; suppression of dust entrainment and reduction of emissions of sulfur dioxide into environment.

INTRODUCTION

According Pyrometallurgical processes are the major links of production of copper, nickel, and associated metals (Schlesinger, 2011). Autogeneous processes on the basis of chemical energy of sulfide ores and various concentrated products are the preferred approach to development of pyrometallurgy. Potentials of autogeneous processes are not implemented completely, which is confirmed by general features peculiar for all autogeneous assemblies:

1. Air blasting per unit of operation volume is restricted by occurrence of bulk emissions and ranges from 5 to 14 $m^3/(min \cdot m^3)$ for various alternatives of air blasting (Shalygin, 2003; Shalygin, 2006).

2. Increased content of magnetite and non-ferrous metals in slags of autogeneous smelting evidences incomplete reactions of slag generation upon formation of final products (Schlesinger, 2011; Shalygin, 2006; Sridhar, 1997).

3. Non-stable pattern of gaseous dynamics in furnace and beyond promotes generation of hard melting skulls, preventing recycling of dust gas mixture (Greiver, 1999; Krupnov, 2013);

4. Isolated positions of heat generation and heat exchange lead to forced heat losses with cooling elements and decrease in campaign lives (Shalygin, 2003; Seregin, 2001);

5. Autogeneous assemblies require for additional power sources, which is accompanied by average unrecoverable heat losses into environment in amount of 15-20% (in terms of total input) (Krivandin, 2002; Lisienko, *et al.*, 2005).

In this or that extent these interrelated critical properties are characteristic to Russian autogeneous processes (PV, KIVCET, AAP, and others) (Lisienko, *et al.*, 2005; Reznik, *et al.*, 2003; Vanyukov, 1988; Bulatov, 2014), especially pronounced in decreased quality of processed ore and involvement of low exothermic and hard melting products of metallurgical process (Eroshevich, 2012; Boduen, 2014).

As a consequence of the researches by Shalygin a radically innovative jet swirling apparatus (JSA) was

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proposed (Shalygin, 2003; Shalygin, 2006; Shalygin, et al., 2006), its main concept is possibility of multiple increase in ultimate amount of air blasting per unit of operation space of apparatus, provided that intensive regular heat and mass exchange in molten bath is maintained created by spatially oriented air blasting.

EXPERIMENTAL PROCEDURE

Based Theoretical studies were based on known physicochemical regularities of processes occurring at the stage of oxidizing pyrometallurgical processing of sulfide polymetallic raw stuff. Mathematical simulation was performed using MathCad and ANSYS software. Experimental studies were performed on cold and fire models in laboratories of Mining University, on large scale pilot facilities at OAO Kola MMC. Chemical composition was determined using X-ray spectral analysis. Composition of gaseous phase was determined by chemical analysis. Technical measurements were performed using regular thermocouples, thermal camera, gauges equipped with disk diaphragms.

INSIGHTS RESULTS AND DISCUSSION

The main task of pyrometallurgical processes is consecutive removal of iron sulfide from complex sulfide system. Oxidation of iron sulfides is accompanied with heat release, hence, the possibility of specified autogeneous pyrometallurgical process in an apparatus of certain design is revealed by solution of extended equation of heat balance (Shalygin, 2003):

$$X = \sum Q_{prod} + Q_{ext} + Q_{end} - Q_{blend} - Q_{blast} - Q_{ex} \text{ kJ/t} \quad (1)$$

where X is the heat consumption by blend; $\sum Q_{prod}$ is the heat content of process product; Q_{ext} is the external heat loss; Q_{end} is the heat of endothermic reactions; Q_{blend} is the heat content of initial blend; Q_{blast} is the heat content of air blasting;

Q_{ex} is the heat of exothermic reacting considered in material balance.

Remarkable attention in solution of minimization of power consumption and achievement of autogeneous essence of pyrometallurgical facility should be paid to its external heat losses Q_{ext}. External heat losses were determined using thermal imaging of main structures of several pyrometallurgical facilities Table 1 involved in technological flowchart of copper production.

On the basis of performed measurements total amount of heat losses into environment was determined per one ton of processed blend, taking into consideration peculiar features of pyrometallurgical assembly and its dimensions:

Table 1. Temperature distribution over external surface of metallurgic assemblies of copper production process flow (matte smelting–conversion–fire refining)

No.	Item		t_{max}, °C	t_{min}, °C	t_{avg}, °C
	Vanyukov smelting furnace (matte smelting)	Slag end	159	47	93
			153	94	132
		Tuyere water jackets	52	49	52
			51	49	50
		Brickwork	52	47	49
			58	46	56
		Water jackets	46	42	44
			40	35	39
		Uptake	41	35	40
			205	156	172
		Roof	211	124	174
			213	114	180
		Uptake	202	165	182
			84	70	76
		Matte mixer	211	156	167
			190	155	183
	Horizontal converter (conversion)	External wall	326	269	309
			333	201	296
		Hood of tuyere area	230	211	220
			249	184	231
21.	Anodic furnace (fire refining)	Lining	283	237	257
22.			375	119	290

$$Q_{ext} = \left(17710 \cdot F_{surf} + 786000 \cdot \mathrm{F_{hole}} + 84 \cdot \mathrm{W_{cool}} + 84 \cdot \mathrm{W_{lance}}\right) \cdot \tau_{\mathrm{bal}} \quad \mathrm{kJ/t} \quad (2)$$

where F_{surf} is the uncooled surface area of the apparatus with account of ribbing, m²; F_{hole} is the surface area of open holes, m²; W_{cool} is the water consumption for cooling of water jackets, kg/t; W_{tuyere} is the water consumption for tuyere cooling, kg/t; τ_{eq} is the balance time, h.

The equilibrium time is a function of specific performance of pyrometallurgical apparatus and external surface area of furnace:

$$\tau_{bal} = 24 / \mathrm{a} \cdot \mathrm{F} \text{ , h} \quad (3)$$

where a is the specific performance, t/m²·day, F is the surface area of representative cross section of furnace, m².

Hence, the balance time as a factor determining heat losses into environment at preset daily performance, other conditions being equal, is inversely proportional to specific performance and representative dimensions of the apparatus.

Specific performance of pyrometallurgical apparatuses

Based on the principle of forced supply of gaseous oxidizer, depends undoubtedly on amount of air blasting supplier per unit time. Possibility to increase performance of autogeneous apparatuses is limited by physical impact of blasting jets on molten medium. Combination of dynamic head, forces of thermal expansion of gases in the melt, and buoyancy forces of gas bulk ascending from the melt result in occurrence of massive melt ejection from operation space of the apparatus. Method of air blasting stipulates certain ultimate blasting load. Upon experiments on models and industrial facilities the following values of ultimate blasting load were determined per 1 m³ of operation space of apparatus (Table 2) (Shalygin, 2003; Shalygin, 2006; Shalygin, 2006). Possible increase in blasting load and, hence, in specific performance will significantly influence on the structure of heat balance of the apparatus, which in particular will expand possibilities of autogeneous oxidizing processing of sulfide polymetallic raw stuff of non-ferrous metallurgy.

Reduction of external heat losses due to increased performance can compensate certain decrease in heat of exothermic reactions related with processing of ore with decreased sulfur content and provide processing of such ore in autogeneous mode without additional supply of any fuel.

Heat and mass exchange of jet swirling apparatus

Separated positioning of heat generation and heat exchange areas leads to thermal overloads of furnace lining near local sites of intensive heat release and decrease in campaign lives (Table 3).

Air blasting procedure into melt via tuyeres located in copper water jackets of Vanyukov smelting furnace cannot be considered as successful, which is evidenced by short operation lifetime with air blasting according to observations in 9 months (Table 4).

The main reasons of destruction of water jackets were the factors of chemical corrosion with generation of sulfur containing copper compounds decreasing mechanical strength and heat conductivity as well as thermal, caused by variations in composition of exothermic blend and breakage of skull protection.

Regular ordered mass transfer stipulates uniformity

Table 2. Ultimate specific blasting load on various units of autogeneous type reduced to normal conditions (T = 273 K, P = 101315 Pa)

Autogeneous unit	Air blasting type	Blasting load, m³/(min·m³)
Bessemer converter	Bottom distributed	4-5
Vertical oxygen converter	Upper non-submerged	6-8
Kaldo converter	Inclined non-submerged	6-9
Horizontal converter	Side-type submerged unilateral	7,5-13
Vanyukov smelting furnace	Side-type submerged bilateral	8-10
Jet swirling unit	Spatially oriented	50-85

Table 3. Performances of autogeneous units

Autogeneous apparatus	High temperature area	Temperature in heat generation area, °C	Campaign lives, days
Vertical oxygen converter	Blasting crater	> 2500	330
Horizontal converter	Near-tuyere area	1500-1750	60
Vanyukov smelting furnace			180-330
Suspension smelting	Reactive blend	1700-1800	330

Table 4. Downtime indices of Vanyukov smelting furnace, Balkhash copper smelter (OAO Kazakhmys)

No.	Failures	Time, hours
1	- scheduled preventive operations	37.58
2	- destruction of water jackets	97.42
3	- power failure	30.17
4	- shutdown of air oxygen mixture supply	27.67
5	- shutdown of gas exhausting system	90.42
6	- technological and transport shutdowns	31.33
7	- major overhaul	974.00
Total		1288.59
Calendar period of observations -		6288
Coefficient of operation with blasting without accounting for major overhaul period -		0.95
Coefficient of operation with blasting in calendar period -		0.80

of thermal and chemical fields in the melt bulk and provides the most favorable conditions for technological process.

Mathematical interpretation of swirling effect of liquid as a consequence of application of tangential stresses to its surface:

$$\upsilon_{unit}(r,z) = -2\cdot\sum_{k=1}^{\infty}\frac{q_k}{J_0(\beta_k)\cdot\beta_k}\cdot\frac{sh(\beta_k z)}{ch(\beta_k h)}\cdot J_1(\beta_k r) \quad (4)$$

assuming in the first approximation that the tangential stresses on free surface are concentrated in narrow ring with the radius $R_0 = r_0 \cdot R$ and width $\delta << R$, then

$$q_k = \frac{1}{2\delta}\cdot r_0\cdot J_1(\hat{a}_k r_0) \quad (5)$$

Substituting equations (4) and (5) in stationary mode we obtain the velocity field:

$$\upsilon_{unit}(r,z) = -\frac{1_0}{\pi}\cdot\sum_{k=1}^{\infty}\frac{J_1(\beta_k r_0)}{J_0(\beta_k)\cdot\beta_k}\cdot\frac{sh(\beta_k z)}{ch(\beta_k h)}\cdot J_1(\beta_k r) \quad (6)$$

where r_0 = R_0/R, h= H/R are the dimensionless geometrical characteristics of cylinder; R is the radius, m; R_0 is the radius of application of tangential stresses, m; H is the height of liquid layer, m; β_k is the zero of Bessel function, υ is the relative velocity.

Using the MathCad mathematical model the maximum velocity of liquid swirling was determined near intensive physiochemical interactions (area in the vicinity of bath surface) at the radius r_0 of application of tangential stresses to its surface equaling to 0.7 (Fig. 1) which corresponds to the results of physical simulation (Konovalov, 2002).

Experimental validation of the JSA concept in melt was performed for various compositions of copper nickel material in the course of fire experiments in the range from 1.0 dm³ to 1.0 m³ and specific blasting load from 26 to 90 m³/min·m³ (Table 5).

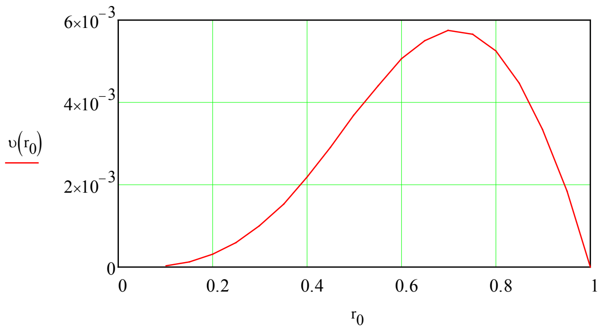

Fig. 1 Dependence of dimensionless rate in z and r coordinates at h = 0.9.

The pattern of steady process was visually observed upon intensive swirling of the melt (matte, copper and nickel concentrated products from converter matte plant) and without emissions. Blowing process and content of sulfurous anhydride (5-12% upon air blasting) in gases confirmed possibility of their efficient recycling, herewith, cyclone effect was observed above the melt preventing emissions of melt and dust.

Therefore, large scale experiments made it possible to obtain positive response to initial question concerning possibility of non-submerged blasting by means of high pressure spatially oriented jets to create ordered mass and heat transfer, meeting the requirements of kinetics of physicochemical interactions in heterogeneous system.

Radial axial tuyeres of new design

Peripheral spatially oriented air blasting is implemented by means of new design of tuyere, its development was based on the following required conditions ((Konovalov, 2015; Shalygin and Konovalov, n. d.):

– Possibility of multiple reduction of dimensions of tuyere fire zone and decrease in water consumption for its cooling;

Table 5. Fire experiments in the mode of jet swirling apparatus

No.	Material	Content, %					Blasting unit	Volume m^3	Tip diameter, mm	Specific blasting load, $m^3/(min \cdot m^3)$
		Cu	Ni	Fe	S	SO_2				
1	Cu-Ni matte	6.0	10.5	48.3	28.6	9-12	6 mini-tuyeres	0.001	1.6	50
2		8.3	13.0	35.0	N/A	5-11.5	6 tuyeres	1	8	26
3	Cu concentrate	69.2	3.7	3.5	20.5	11-12	6 mini-tuyeres	0.001	1.6	65
4		68.7	3.4	4.2	21	10-12	unit (6 tips)	0.001	1.6	90
5		68.7	3.4	4.2	21	N/A	unit (6 tips)	0.02	4	60
6	Ni concentrate	2.7	72.8	4.8	17.6	N/A	6 tuyeres	0.03	4	35

– Cooling under depressurizing;

– Possibility to decrease the height in case of horizontal positioning of several tuyeres;

– Creation of regular swirling motion of melt.

Such requirements are met by radically new tuyere design (Fig. 2).

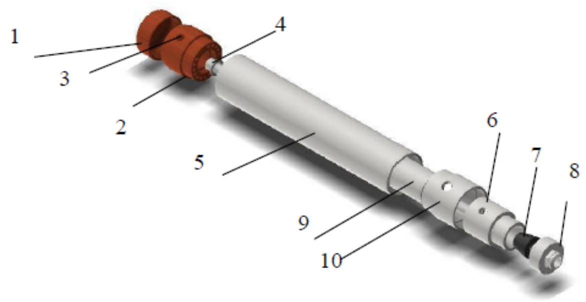

Fig. 2 Design of radial-axial tuyere: 1 – cap, 2 – tuyere head, 3 – blasting tip, 4 – central tube, 5 – external tube, 6 – blasting cartridge, 7 – seal, 8 – counter nut, 9 – intermediate tube, 10 – water cooling cartridge.

Analytical mathematical description of channel of complex shape is difficult, hence, the coefficients of hydraulic resistance were determined using experimental bench for hydrodynamic tests (Shalygin, *et al*., n. d).

Experimentally determined hydraulic resistance of the tuyere cooling system was significantly lower than 1 atm which proves the validity of previous assumption of possibility to apply explosion-proof cooling under depressurizing.

Heat loads on tuyere were calculated in ANSYS software (ANSYS CFX-Solver Theory Guide ANSYS, Inc. 2009) using SST model (Shear stress transport) (Menter, *et al*., 2003). As a consequence, temperature distribution of cooling agent was determined. It is established that under extreme conditions of heat transfer water consumption to tuyere with tip diameter of 20 mm should be about 0.018 m^3/s.

Skull protection of lining is the most efficient protection against chemical and thermal destruction of operation surface of pyrometallurgical apparatus contacting with molten medium: both of brickwork and of metal surface.

Contrary to the existing variants of jet air blasting (vertical non-submerged, horizontal submerged) the proposed approach to creation of spatially oriented non-submerged jets distributed over periphery of cylindrical body of the apparatus and creating controlled swirling of melt bulk due to tangential stresses is the most favorable also for formation of steady skull.

Such mode was preliminary estimated using specialized laboratory facility (Shalygin, 2003). The estimation was performed for qualitative comparison of heat exchange conditions between melt and wall upon vertical air blasting by single tuyere and by JSA. Both experiments were performed maintaining similar air pressure and flow rate, the same temperature, the same mode of water supply. The pattern of skull generation was obviously different. Upon vertical air blasting skull is heterogeneous in thickness and structure at various areas of the water jacket. Either it is completely absent, or it is rather thick. This can be attributed to already mentioned randomness of behavior of blasting crater, formed by vertical jet, its oscillations, displacements in transversal direction. In addition, significant vertical melt splashes are observed, emitted above the model body, upon single vertical air blasting. When air blasting is performed in the mode of JSA, vertical splashes of the melt are nearly absent, and skull is distributed over total surface of jacket wall as uniform dense layer.

Melt swirling under the impact of spatially oriented jets creates ordered mode of convective thermal action of melt onto cylindrical apparatus wall. Temperature of the swirling melt contacting with the wall due to bath mixing satisfies certain averaged value, which is significantly below the temperature developed in the area of blasting craters. At sufficient heat conductivity of refractory wall arrangement of water cooling jackets in it can provide generation of steady skull on fire surface, which can promote long operation lifetime at high thermal intensity characteristic to autogeneous process.

Cyclone effect of gaseous phase above melt. On the basis of experiments with cold and fire models of JSA the cyclone effect, visually observed in gaseous phase above melt, was mathematically simulated using ANSYS CFX package. Total hydrodynamic characteristic of JSA is illustrated in (Figs. 3–5) by calculations using control volumes.

CONCLUSIONS

The Physicochemical concept of thermal balance is developed in detail, enabling estimation of each item of thermal balance and evaluation of possibility of reasonable control of these items.

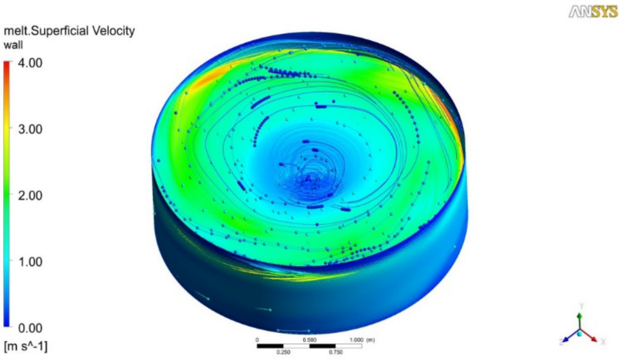

Fig. 3 Behavior of liquid bath.

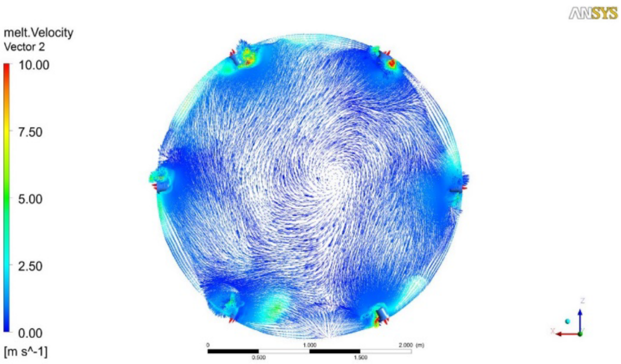

Fig. 4 Dynamics of gaseous phase at the height of radial axial tuyeres.

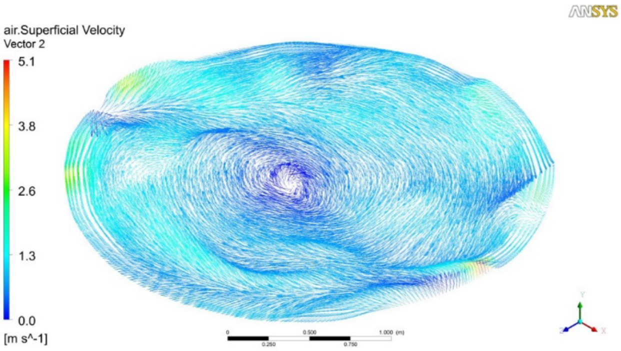

Fig. 5 Dynamics of gaseous phase above radial axial tuyeres.

Interrelation between external heat losses and specific performance of metallurgical apparatus is demonstrated. The factor, which restricts increase in specific performance, is the method of air blasting.

In order to perform autogeneous processing of polymetallic raw stuff in forced mode a radically innovative, spatially oriented air blasting was proposed enabling combining of areas of heat generation and exchange in operation space of autogeneous assembly to the greatest possible extent.

Experiments with cold models and fire assemblies confirmed possibility to increase specific blasting load in two times and higher in comparison with existing methods of air blasting.

Flow dynamics pattern in the proposed apparatus of jet rotation under the action of tangential stresses created by kinetic energy of distributed spatially oriented jets is established.

A new design of autogeneous assembly for processing of sulfide polymetallic raw stuff is proposed on the basis of spatially oriented jets creating swirling motion of melt in overall apparatus space.

A radically new design series of tuyeres is developed, which provide generation of ultrasound jets outflowing at any preset angle to bath surface.

The tuyeres, possessing very moderate hydraulic resistance of cooling channels, correspond to conditions required for underpressure cooling system, and water consumption for cooling of set of 6 tuyeres is lower by three times in comparison with vertical single tuyere.

Possibility to create steady skull protection under regular controlled heat and mass transfer in molten bath occurring under the action of ultrasound spatially oriented jets.

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