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Mathematical models based analysis of the efficiency of joint processing of pulverized coal and metallurgical waste using a melting gasifier

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Abstract. The paper presents options for almost waste-free technologies for joint and separate processing of pulverized metallurgical and coal waste based on a jet-emulsion melting gasifier with obtainment of three liquid products. The results of mathematical modeling are presented, technological modes and main design parameters of the units are determined.

1. Introduction

In previous works [1, 2], two versions of melting gasifiers based on a jet-emulsion metallurgical reactor [3] were presented, which allow, along with the direct reduction of iron oxides, synthesis gas to be obtained, as well as a modified version of the gasifier intended for processing waste coal without iron-containing additives.

The presented work focuses on the problem of mathematical modeling of several possible designs and technologies for joint as well as separate processing of pulverized iron-containing and coalcontaining wastes based on specific examples of the design of pilot gasifier.

2. Mechanism of the process and chemical transformations

Figure 1 shows a diagram of a gasifier based on a jet-emulsion metallurgical reactor. The technology of the process carried out in this gasifier (protected by the patent) was discussed in detail earlier [4]. Here we provide a fairly visual diagram of this technology without detailed explanations, limiting ourselves only to a brief description of the designations.

We hope that on the basis of the previously presented description [4] and the symbols shown in figure 1, the process of functioning of this scheme becomes quite clear, especially since below there is a diagram of chemical transformations in individual zones of the unit (figure 2), referring to, we will consider the processes taking place in each of the zones using a specific example.

The combustion of fuel in the reactor-oscillator 2 is carried out with a lack of oxygen to CO, heating and partial reduction of oxides also occur here, but the main task solved in this reactor is the creation of a gas suspension. Most of the reduction processes are completed in the column reactor 5 (figure 1). Iron reduced from oxides is lowered along the periphery of the column reactor into the storage chamber 7. Here, partial post-combustion of carbon monoxide to dioxide is carried out. In this case, post-combustion occurs inside the gas-slag emulsion by supplying oxygen. The total oxygen consumption is selected in accordance with the heat balance. To balance the process with respect to temperature, the degree of CO post-combustion is changed by changing the amount of gaseous oxygen

or steam. In this case, priority is given to obtaining metal and slag of a given composition. The sequence and relationship of chemical transformations is clear from figure 2.



Figure 1. Waste-free gasifier based on jet-emulsion reactor: 1–gas-tight loading device; 2–reactoroscillator; 3–connecting channel with gas-dynamic self-locking; 4–column order-emulsion reactor; 5 –gravity separator for metal, slag and gas; 6–gas dynamic cushion separating the upper and lower parts of the column reactor; 7–metal in the forehearth; 8–skull cooling; 9–connecting channel; 10– auxiliary channel for maintaining the level of gas suspension above channel 9; 11 – slag receivers; 12 –drum granulator; 13 –lattice; 14 –gas reformer, 15 –coke or adsorbent layer.



Figure 2. Scheme of chemical transformations.

3. Mathematical modeling of combined processing of metallurgical and coal waste

A variant of a specific implementation of the process is considered by the example of direct reduction of iron with simultaneous production of synthesis gas in a pilot unit with an ore capacity of 1 kg/s or 3.6 t/h [3, 4]. The technology is implemented in the unit shown in figure 1.

With the help of the "AutoAstra" instrumental system [5, 6], thermodynamically acceptable conditions and compositions of metal, slag and gas were obtained for iron ore consumption of 1 kg/s. At a coal consumption of 0.674 kg/s and an oxygen consumption of 0.382 kg/s, a metal was obtained with a carbon content of 0.52% and a temperature of 1873 K, as well as an exhaust gas in an amount of 1.06 kg/s [4], the composition of which is given in the table 1.

Table 1. Chemical composition of the gas.									
Content, %	СО	CO_2	H_2	H_2O	N_2	Total			
By mass	92.83	3.21	2.14	1.74	0.02	1.06			
By volume	72.78	1.60	23.49	2.13	0.01	1.41			

This gas, in close contact with the slag (in the form of a gas-slag emulsion) through channel 9

(figure 1), enters the slag receiver 11, where steam is supplied towards the slag flow in order to cool and granulate the slag. At the same time, steam-gas correction reactions occur simultaneously

$$CO + H_2 O = CO_2 + H_2; (1)$$

$$CO_2 + C = 2CO; \tag{2}$$

$$H_2 O + C = C O + H_2, (3)$$

as a result, the H_2 /CO ratio increases, and the temperature of the gas coming from the column reactor 5 through channel 9 decreases from 1500 °C to 800 °C. Thus, a kind of chemical recovery of the physical heat of the waste gases is obtained. This gas through the grate 13 enters the coal (coke) layer into the chamber 14.

The subsequent operations for correcting the gas composition are carried out in chamber 14. In this layer and above it, reactions (1) - (3) proceed in the conditions of excess carbon.

Table 2 shows variant calculations of gas correction as a result of its interaction with coal and steam.

Control actions						Volumetric composition of gas, %					
	Coal	Steam		Gas						Ratio	
No.	consumption,	consumption,	Т, К	mass,	CH_4	CO	CO_2	H_2	H_2O	CO/H_2	
	kg/s	kg/s		kg/s							
1	0.20	0.30	1100	1.53	0.42	60.47	3.18	34.11	1.82	1.77	
2	0.25	0.375	1065	1.65	0.62	56.16	5.08	35.27	2.87	1.59	
3	0.30	0.45	1042	1.73	0.81	52.32	6.79	36.21	3.87	1.44	

Table 2. Variants of calculating the gas chemical composition.

Since the coal in the layer is in excess, the control is carried out by changing the amount of supplied steam. As it can be seen from table 2, in the third correction option, it was possible to increase the H₂ content from 23.49 to 36.21%, and the CO content was reduced from 72.78 to 52.32%.

Further movement in this direction (correction due to coal and steam) becomes ineffective, because, due to a decrease in temperature in chamber 14, the content of CO_2 and H_2O increases.

It should be noted that the obtained gas (line 3 in table 2) is close in composition to the synthesis gas obtained by the Fischer-Tropsch method [7] in the Winkler fluidized bed gasifier: CO 48.2%; H₂ 35.2%; CO₂ 13.8%; CH₄ 1.8%; N₂ 0.9%; as well as in the Koppers-Totzek gasifier, which processes coal dust [8, 9]: CO 57.2%; H₂ 30.7%; CO₂ 10.5%; CH₄ 0.1%; N₂ 1.2%; H₂S + COS 0.3%.

As it can be seen from the comparison of the above compositions of the crude synthesis gas with the one obtained by us above (row 3 in table 2), in terms of the content and ratio of CO/H₂, all three gases are close, but the gas we obtained has almost half the content of carbon dioxide, which requires

significantly lower costs for cleaning and increases the yield of the final product. But, the most important advantage is that in this case, synthesis gas is essentially a by-product, while in the known technologies for producing motor fuel from coal, up to 70% of capital costs are related to the production of synthesis gas, and in the known gas generators 20–25% of the fuel is spent on maintaining the temperature in the gasification zone [7–9]. In the example considered by us, even the thermal energy of the waste gas is converted (regenerated) with the chemical energy of the synthesis gas.

Due to the lower supply of the working mixture to the column reactor, a high layer of foamed slag is maintained in it, inside of which pulverized coal is combusted to carbon monoxide, which creates conditions for the full use of the energy of the initial fuel. And due to the creation of high turbulence in the column reactor, connecting channel 9 and slag receiver 11, gas-saturated slag foam is broken into small aluminosilicate granules and microspheres [10], which are very liquid on the market and cost more than metal (from 35 to 50 thousand rubles. per tonne). Thus, a practically waste-free production is obtained.

The resulting gas can be burned directly in the stream in a piston mini-power plant [11]. Such power plants are serially produced [12].

If necessary, this gas can be converted into motor fuel [13, 14]. In addition, a certain amount of metal accumulates in the forehearth 7. Thus, the polluting waste turns into three valuable liquid products. When gas is burned in a piston power plant, a completely non-volatile technology scheme is created, including the production of its own oxygen and nitrogen [1, 11].

4. Melting gasifier for processing waste coal

If the task is to process only coal enrichment wastes (without the possibility of adding iron ore materials), the problem arises of maintaining a stationary oscillatory mode of the reactor-oscillator 2 (figure 1), since the content of the condensed phase in the gas suspension ejected through the gasdynamically locked channel 3 sharply decreases. In this regard, it was decided to somewhat simplify the unit and abandon the use of an oscillator reactor. But this naturally raises the question of how to ensure the main condition for the implementation of the principle of self-organization [15, 16] - a large deviation from thermodynamic equilibrium, and stable maintenance of the slag foam in the column reactor without a sufficient amount of iron oxides in the slag.

It is advisable to show the ways of solving this problem directly when describing the operation scheme of the modified version of the correct gasifier shown in figure 3.

The mechanism of the process in this unit is implemented as follows. In the induction heated forehearth 1, a certain amount of metal is pre-melted. Further, during the operation of the unit, the surface of this metal is blown by hard jets of oxygen, as a result of which a zone of highly oxidized slag 2 is created above the metal, since the reactions of direct oxidation of carbon with the formation of FeO and Fe_2O_3 are intense. Particles of coal fall on this zone from above, as a result of which zone 3 is formed above this zone, where the reduction reactions of these oxides are intensively proceeding:

$$(Fe_2O_3) + C_s = 2(FeO) + \{CO\},$$
 (4)

$$(FeO) + C_s = [Fe] + \{CO\}$$
(5)

and there is an intense release of carbon monoxide. As a result, a flow of bubbles of carbon monoxide is created, which creates intensive mixing and turbulization of the slag emulsion layer. To improve the process of mixing falling coal particles into the emulsion, an additional turbulence zone 4 is created.

This is achieved due to the coaxial arrangement of the steam nozzles, while at the same time steamoxygen conversion of the gas is also carried out according to reaction (3), which makes it possible to increase the calorific value of the commercial gas by using the physical heat of the system.



Figure 3. Melting gasifier for processing waste coalent.

Post-combustion of fine carbon particles in the upper part of the column reactor is carried out using an oxygen lance 5. The resulting gas suspension under pressure in the column reactor is discharged through channel 6 into a slag receiver, where slag particles are also turbulized and cooled using coaxially located steam nozzles. The resulting closed and open microspheres, as well as strongly foamed shapeless slag particles, are delivered to the conveyor, then gravitational separation and sieve screening are carried out.

The gas cooled in the receiver is fed through a grate with an adsorbent layer on it to a wet or dry gas purifier, and then to a piston engine, which generates electricity used, among other things, to obtain oxygen and nitrogen, and the bulk of the energy is sent to the network.

Thus, the above problem of gasification of coal preparation wastes is solved without using metalcontaining wastes, but it should be emphasized once again that the possibility of using these wastes makes it possible to increase the efficiency, including the technology discussed above.

5. Simulation results of processes in the melting waste gasifier

For this version of the technology, using the "Engineering Metallurgy" tool system [17] and the "Terra" tool system [5], mathematical modeling was carried out in order to determine the optimal flow rates for oxygen and water vapor for a given unit capacity of 1 kg/s (3. 6 t/h) waste coal. The composition of waste coal is shown in table 3.

Table 3. Composition of waste coal.								
Moisture	Ash	Organic part	Volatile					
16.44 %	30.00 %	52.00 %	18.00 %					

As a result of solving the optimization problem, the flow rates of oxygen and water vapor were determined, which ensure the combustion of coal preparation wastes and the heating of combustion products to a temperature of 1500 °C. The flow rates were 0.624 and 0.061 kg/s for oxygen and water vapor, respectively.

The material balance of the process is shown in table 4.

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I able 4. Material balance in the main unit.								
Input, kg/s		Consumption, kg/s						
Waste from coal preparation	1.00	Gas	1.452					
Oxygen	0.624	Slag	0.212					
Steamp	0.061	Metal	0.022					
Total	1.685	Total	1.685					

The amount of iron reduced in the main unit was 0.022 kg/s, and the amount of slag thrown into the slag receiver was 0.212 kg/s. The pressure in the main unit was 4.3 atm.

The calculated composition of slag foam and gas is presented in tables 5, 6.

Table 5. Slag foam composition in the main unit.										
Composition	SiO ₂	Al_2O_3	FeO	CaO	MgO	P_2O_5	S	Na_2O	K_2O	Total
%	59.477	25.699	1.358	2.469	2.469	0.000	3.928	2.132	2.469	100.0
kg/s	0.142	0.061	0.003	0.006	0.006	0.000	0.009	0.005	0.006	0.238
Table 6 . Composition of the gas phase in the main unit.										
Composition		СО	CO_2	N_2	H_{2}	2	H_2O	SO_2	t, °C	
Balance calculation		5	9,55	0,31	0,03	26,47		7 12,61		1500
Calculation b	oy "Terra"	5	5,46	4,60	0,69	30,	01	9,22	0,01	1500

Modeling of processes in the slag receiver. The gas-slag emulsion formed in the main unit enters the slag receiver, where it is cooled with water vapor to a temperature of 750 °C. Cooling occurs, inter alia, as a result of steam gasification reactions (1), (3).

The study of the conditions of these reactions was carried out by the method of thermodynamic modeling using the "Terra" software package.

As it can be seen in figure 4, the most interesting area in which it is possible to choose the optimal ratios of the concentrations of CO, H₂ and CO₂ is in the temperature range 650 ÷ 850 °C. This conclusion is also confirmed by figure 5, which shows the dependence of the CO and H₂ concentrations on temperature.

As a result of solving the optimization problem, the flow rates of steam and coal preparation wastes required for cooling the gas in the slag receiver to 750 °C were obtained. Steam consumption was 0.068 kg/s, coal preparation waste -0.233 kg/s. The composition of the gas phase in the slag receiver is shown in table 7 (line 1). Rows 2 and 3 of this table are given to compare the compositions of gases in the previously mentioned Winkler and Koppers-Totzer gasifiers [7-9].



Temperatre, ⁰C

Figure 4. Dependence of gas composition on temperature in the main unit.



Т, К

Figure 5. Dependence of CO and H₂ concentrations on temperature in the slag receiver.

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Table 7. Composition of the gas phase in the slag receiver.									
Composition, vol. %	СО	CO_2	N_2	H_2	H_2O	SO_2	T °C		
Calculation by "Terra"	50.32	9.08	34.24	0.64	4.70	0.14	750		
Koppers-Totzer	57.2	10.5	30.7	1.2	-	0.3	1500÷1600		
Winkler	48.2	13.8	35.2	0.9	-	-	900÷1200		

But once again we emphasize that in our case, in addition to gas, we will get highly porous commercial slag and metal.

6. Gasification process with obtaining high quality synthesis gas

Due to the fact that the unit discussed above has a small specific volume, it can be made in a mobile version and located near the waste generation or mines where methane is extracted from the beds.

In this case (figure 6), it becomes possible to bring the composition of the synthesis gas obtained above by the steam-oxygen conversion method (table 7, line 1) to a very high condition that fully satisfies the stoichiometric ratio. For example, for dimethyl ether (synthetic diesel fuel) in accordance with its stoichiometric formula $CH_3 - O - CH_3$, the optimal CO/H_2 ratio is 2/3. Table 8 shows the results of the correction in the chamber 8 of the gas composition taken from table 7 by oxygen and temperature conversion of methane by reactions

$$CH_4 + \frac{1}{2}O_2 = CO + 2H_2, (6)$$

$$CH_4 = C + 2H_2. \tag{7}$$

First, the gas composition is corrected by oxygen reforming of methane supplied to chamber 8 according to reaction (6).

As can be seen from table 8 (options 1 and 2), the H_2 content in the resulting gas mixture increases with a simultaneous increase in the mixture temperature.

As can be seen from the second version of the thermodynamic calculation, a synthesis gas was obtained with a volume ratio of CO/H_2 close to 1/1, more precisely 45.20/45.80, which can already be considered a good result, since a high-calorific gas was obtained, which can be used as energy fuel, and as a raw material for synthetic fuels.

However, with such a gas composition, synthetic fuel in the catalytic synthesis unit will be obtained in smaller quantities and of worse quality (due to dilution with unreacted ballast components)

than it is possible if the optimal stoichiometric CO/H_2 ratio of 2/3 is observed. Considering that there is a certain margin of gas temperature (1055 K), a decision is made to reach the final stage of correcting the gas composition due to the temperature reforming of methane according to reaction (7).

Table 8. Gas chemistry correction.													
	Control actions								Volumetric composition of gas, %				
No	Coal consump -tion, kg/s	Steam consump- tion, kg/s	Methane consump- tion, kg/s	Oxygen consumption, kg/s	Т, К	Gas mass, kg/s	CH ₄	CO	CO ₂	H_2	H ₂ O	Ratio <i>CO/H</i> ₂	
1	0.3	0.45	0.20	0.20	1050	2.07	1.10	47.08	4.53	42.80	3.55	1.1	
2	0.3	0.45	0.25	0.25	1055	2.15	1.15	45.20	4.25	45.80	3.60	1	
3	0.3	0.45	0.45	0.25	1010	2.15	2.40	34.00	5.80	51.50	6.22	2/3	

As can be seen from the comparison of options 2 and 3 (table 8), the solution to the problem is achieved by increasing the methane consumption by 0.2 kg/s, without increasing the oxygen consumption. At the same time, a gas was obtained with a volumetric content of CO and H₂, respectively, 34.0 and 51.5%, which corresponds to a CO/H_2 ratio of 2/3.



Figure 6. Gasifier for obtaining high quality synthesis gas.

Thus, in the case of installing a melting gasifier near the place of methane extraction, synthesis gas with an optimal ratio of CO and H_2 can be obtained, which will create conditions for obtaining highquality motor fuel with high economic efficiency.

7. Conclusion

Designs of melting gasifiers and technologies for processing dust-like fractions of coal enrichment with and without additives of metal-containing waste are considered. Mathematical modeling of three technology options was carried out, the optimal consumption of materials in the main unit and slag receiver was determined, which ensure the specified process parameters: temperature and composition

of the gas phase, provided that three liquid products are simultaneously obtained – metal, highly porous slag and synthesis gas.

It is shown that by steam-oxygen conversion in the slag receiver, synthesis gas can be obtained, essentially as a by-product, of somewhat better quality than in the known special gasifiers, where to obtain the required temperature it is necessary to additionally burn 20-25% of coal.

References

- [1] Tsymbal V P, Rybenko I A, Olennikov A A, Rybushkin A A, Kozhemyachenko V I and Sechenov P A 2018 *IOP Conf. Series: Earth and Environmental Science* **206** 012048
- [2] Tsymbal V P, Rybenko I A, Kozhemyachenko V I at al 2019 Science-intensive Technologies for the Development and Use of Mineral Resources: Scientific Journal **5** 478–82
- [3] Tsymbal V P, Mochalov S P, Rybenko I A et al 2014 SER Process *Metallurgical Jet-emulsion Reactor* (Moscow: Metallurgizdat)
- [4] Tsymbal V P, Mochalov S P, Rybenko I A et al 2009 Pat. of the Russian Federation No. 2371482 appl. 12.03.2007, publ. 10.27.2009
- [5] Siniarev G B, Vatolin N A and Trusov B G 1982 Computer Application for Thermodynamic Calculations of Metallurgical Processes (Mosow: Nauka) p 260
- [6] Klimov V IU et al 2001 Proc. of the All-Russian Scientific and practical conference Modeling, Software and Science-Intensive Technologies in Metallurgy: (Novokuznetsk: SibSIU) pp 435-439
- [7] Schilling G D, Bonn B and Kraus U 1986 *Coal Gasification* (Moscow: Nedra)
- [8] Falbe E 1980 Chemicals from Coal in tr. From German (Mosow: Khimiya) p 616
- [9] Altshuler V S 1976 New Processes for Gasification of Solid Fuels (Mosow: Nedra) p 280
- [10] Ismagilov Z R, Shikina N V, Zhuravleva N V et al 2015 Solid Fuel Chemistry 4 49-57
- [11] Zaichenko V M and Chernyavsky V M 2010 Academy of Energy 4 58-63
- [12] Rodionova M A, Khrestianovskaia M V and Kukolev M I 2017 Construction of Unique Buildings and Structures 4(55) 50–66
- [13] Terentyev G A, Tyukov V M and Smal F V 1989 *Motor Fuel From Alternative Raw Materials* (Moscow: Chemistry)
- [14] Plate N 2004 Science and Life 11 66–68
- [15] Nikolis G 1979 Self-organization in Nonequilibrium Systems (Moscow: Mir)
- [16] Prigozhin I and Stengers I M 1986 Order From Chaos: A New Dialogue of Man With Nature (Moscow: Progress)
- [17] Rybenko I A 2018 Bull. Ferrous Metallurgy 3 42–47