

Fully environmentally closed technology of gasification dusted fractions of coal concentration with restoration of metals and aluminosilicates from cinders

V P Tsymbal¹, I A Rybenko¹, A A Olennikov², A A Rybushkin¹,
V I Kozhemyachenko¹ and P A Sechenov¹

¹Siberian State Industrial University, 42 Kirova street, Novokuznetsk, 654007, Russia

²Tyumen State University, 15a Perekopskaya street, Tyumen, 625003, Russia

E-mail: tsymbal33@mail.ru

Abstract. The project proposes a completely ecologically and energetically closed waste-free technology for gasification of pulverized coal waste in a suspended slag layer, producing a conditioned synthesis gas, simultaneous reduction and recovery of metal from ash and slag, and also obtaining a highly porous aluminosilicate slag (microspheres) suitable as lightweight aggregate, paints and coatings with low thermal conductivity, as well as adsorbent and other applications.

1. Brief description of the project

To solve the problem of a completely ecologically closed separation of coal (waste coal), the jet-emulsion aggregate SER (self-organizing jet-emulsion reactor), previously developed for the direct reduction of pulverized metallurgical wastes and ores, is used [1, 2]. In the first (spherical) reactor of this aggregate, a large interphase chemical interaction surface is created due to the effective sputtering of the incident charge stream (coal, slag-forming) by counter propagating oxygen jets, the gas suspension formed through a gas-dynamically locked channel is fed to the lower part of the vertical cylindrical reactor, where it forms gas-dynamic "failure grating", on which the gas-slag emulsion hangs. Inside this emulsion and incomplete combustion of coal (up to CO) with almost full use of the resulting thermal and chemical energy. Thanks to the organization of the gravity separation mechanism, the products are separated into metal and gas slag, which enters the lattice of the gas reformer via the slag receiver, where steam-coal combustion and gas enrichment is carried out with hydrogen to a volume ratio H₂/CO equal to 1/1, which is already good the result of [3, 4] for the subsequent synthesis of dimethyl ether is at least better than in the Fisher-Tropsch method, where it is still necessary to separate nitrogen [5].

If there are resources of methane or natural gas nearby, then by oxygen conversion of a relatively small amount of methane (15-20% of the gas volume), it is possible to obtain an optimal H₂/CO ratio of 3/2 for the synthesis of dimethyl ether and 2/1 for methanol, corresponding to their chemical formulas. In this case, it is possible to carry out an almost non-waste catalytic synthesis of motor fuel.

2. The device and principle operation of the SER unit

The process is realized in the aggregate shown in figure 1 as follows [6]. The powdered charge consisting of a mixture of coal (waste) and slag-forming (iron oxides) is fed by a metering feeder 2 to



the central zone of the reaction chamber 1 where a seal disk is formed at the place where the oxygen flows delivered through the inverted directional lances 3 meet. As a result of the dynamic interaction of jets there is an intensive turbulence of the charge flow and the formation of large surfaces for heterogeneous chemical interaction. At the same time, due to incomplete combustion of a part of coal or other reducing agent in the reaction chamber 1, heating and partial reduction of oxides occur in accordance with the proportion of oxygen supplied.

In this unit, which has a certain universality, as a separate task, it is possible to process, that is, gasification, only coal, without the addition of ore or with a minimum addition of ore, only to reduce the process temperature and produce a foamy slag emulsion. With this version of the technology, simultaneously with the incomplete combustion of coal inside the gas-slag emulsion, metals (iron, manganese, etc.) are recovered from the oxides contained in the coal ash and in slag-forming additives. In this case, the composition of the gas by the sum of CO and H₂ approaches 90%.

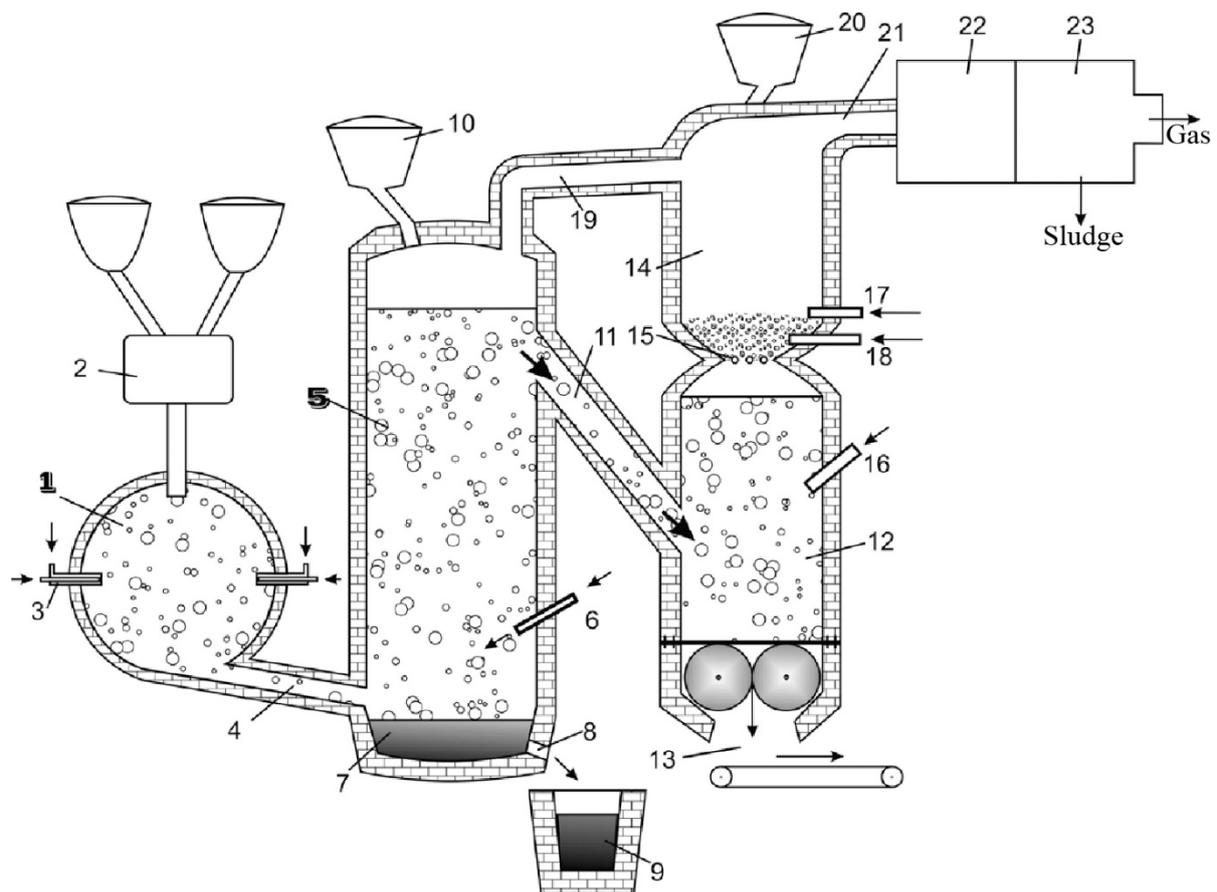


Figure 1. Scheme of wasteless complex gasification.

Technology with the addition of iron oxides (sludge, tailings of enrichment) to coal is very rational, since part of the gaseous oxygen is replaced by oxygen extracted from iron oxides, and the resulting metal or synthesis gas becomes essentially free. At the same time liquid iron and its oxides are simultaneously catalysts, allowing to accelerate the process and increase the yield of some components of synthesis gas.

The use of a vertical column reactor 5 with a lower feed of the reaction gas suspension in combination with a significant deviation of the processes from thermodynamic equilibrium as the refining settler is an important factor that makes it possible to separate the flow of metal deposited in the calculator 7 and the flux of the gas slag emulsion discharged along the inclined channel 11 in a

slag receiver 12 where slag and gas are separated. To bring the composition of the associated process gas from the slag receiver 12 through the cooled grate 15 to the gas conversion chamber 14 to the conditioning composition of the synthesis gas suitable for processing dimethyl ether or methanol under the grate 15, with the continuously expanded material 19 a layer of coke or coal, steam is supplied, and natural gas and oxygen are supplied above the grate 15 to the gas conversion chamber 14, while providing a volume ratio of CO to H₂ in the produced gas 2/3 or 1/2 for production of dimethyl ether or methanol. The resulting synthesis gas from the chamber 14 through the channel 21 is fed to a cooler (for example, a waste heat boiler) 22, a gas purification 23 and then to the consumer, for example, in a catalytic synthesis unit of dimethyl ether or methanol or for energy purposes. The sludge generated in the gas scrubbing 23 is recycled into the process, which allows the production of smokeless technology.

An advantage of the present invention is that the installation in series with the refiner 5 of the gas composition correction chamber 14 combined with the slag receiver 12 and the slag granulator 13 gives a number of additional effects. First of all, there is the possibility of flexible (practically independent) control of the composition of the gas at the outlet from the refining settler 5 and at the outlet from the aggregate as a whole, more precisely from the chamber 14. Combination of the coal gasifier (chamber 14) with the metallurgical unit (reaction chamber 1 and refining settler 5) and the slag receiver 12 allows us to use the physical heat of associated gas and slag to increase caloric content, and the amount of commodity gas. Thus, on the basis of chamber 14, a chemical regenerator (utilizer) of physical heat is obtained. The increased pressure created in the reaction chamber 1 allows the reaction gas to be pushed through all the elements of the unit, which results in a small specific volume of the unit and eliminates the need for using flow and smoke drivers.

If, however, gas is produced in a special gasifier (gas generator), as in the Lurgi-Fischer-Tropsch method, a considerable amount of thermal energy is spent to maintain the temperature (800-1000°C) in the gasification site and nitrogen heating [5], and the gas is obtained low caloric and requires a large expenditure for the separation of ballast components before catalytic synthesis.

3. Characteristics of the products obtained

Due to the fact that the gas suspension weighs in the column reactor 5 and then thrown out at high speed through the inclined channel 11 to the slag receiver 12, has a very high gas content (more than 99%), it is possible to produce highly porous aluminosilicate microspheres. The parameters of the column reactor, the connecting channel and the blast regime are selected in such a way that the so-called inertial-turbulent regime of motion of the two-phase working mixture created in the reactor-oscillator (the first reactor) is fulfilled. In this case, conditions are created when the slag film is "wound" on gas bubbles and hollow spherical structures (microspheres) are formed. Managing the slag regime, it is possible to obtain films of the required chemical composition, and the installation of a gravity separator after the slag-receiver granulator, allows us to classify the microspheres according to the granulometric composition, that is, to satisfy the most stringent requirements. Quality microgranules are more expensive than metal. The following slag composition for microgranules was obtained on the mathematical model (table 1):

Table 1. Chemical composition of microgranules.

Slag	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	Total
%	62.856	26.727	1.412	2.568	2.568	2.217	2.568	100

After the steam conversion, chamber 14 produces a gas of the following composition (table 2), with an H₂/CO ratio of 1/1, that is already suitable for the catalytic synthesis of motor fuel. If such a task is not put, then this gas is fed to a gas turbine or a gas piston machine and turns into electricity, including that used inside the process cycle for the production of oxygen. This completes the first stage of the project implementation.

Table 2. Composition of gas after steam-oxygen conversion.

Gas	CO	CO ₂	N ₂	H ₂	H ₂ O	CH ₄	Total
% By weight	51.518%	31.355%	0.887%	3.680%	12.559%	0.000%	100.00%
% By volume	35.92%	13.91%	0.62%	35.92%	13.62%	0.00%	100.00%

If the task is to convert the synthesis gas into motor fuel, either the gas of the composition obtained above can be used, or by oxygen conversion of not a sick amount of methane is brought to higher conditions corresponding to the chemical formulas of dimethyl ether (diesel fuel) or methanol. Table 3 shows the calculated composition of such a gas.

Table 3. Composition of gas after additional methane-oxygen correction.

Gas	CO	CO ₂	N ₂	H ₂	H ₂ O	CH ₄	Total
% By weight	58.084%	29.434%	0.880%	5.632%	5.970%	0.000%	100.00%
% By volume	35.03%	11.30%	0.53%	47.55%	5.60%	0.00%	100.00%

The metal (iron-carbon melt) located in the casing 8 is necessary for the stable maintenance of the gas-slag emulsion, inside which carbon and volatile constituents of coal are burned. In addition, iron oxide is a catalyst for the formation of hydrocarbons. Periodically produced from a forehearth metal, in the simplest case is sold as a first-class scrap, the market of which is not limited. But, in principle, all metals contained in the coal sol are concentrated in this metal. This task requires separate consideration.

In the presence of a conditioned synthesis gas, the task of catalytic synthesis does not cause any special difficulties, although, of course, it requires additional, but quickly recouped, capital investments. We do not consider this stage of the possible development of the project here.

In the case of the above project, the resulting synthesis gas can be considered as a by-product, since its cost is completely covered by two other liquid products – aluminosilicate micro-granules and metal, as can be seen from table 4, which presents the main technical and economic indicators from implementation of the pilot sample of the unit with a coal output (waste) of 3.6 tons per hour.

The effectiveness of the project is very high: the payback period is less than a year after launch, and every ton of recycled waste (currently polluting the environment) can generate profit of 17 thousand rubles. It should be emphasized that a completely ecologically and energetically closed technological scheme is created, the input stream in which is only coal (waste), yet other products, including oxygen, are obtained within this technology. This allows you to design the unit in a modular mobile version and place it near waste generation sites or in locations remote from electrical networks.

Table 4. Technical and economic indicators.

Input current	Consump., hour	Consump., year	The price of units, rubles/ton, or thousand m ³	Costs, income for the year, million rubles	Notes
Coal (waste), ton	3.6	25920	400*	10.4	
Oxygen, m ³	1075	322500	-	-	Oxygen station (with compressor up to P=150 atm.) – 8 million rubles

Input current	Consump., hour	Consump., year	The price of units, rubles/ton, or thousand m ³	Costs, income for the year, million rubles	Notes
Products received					
Gas, thousand m ³	7.2	51840	4000	207.9	
Al – Si, ton	0.9	6480	35000	226.8	
Metal, ton	0.07	504	18000	9.1	
Possible full generation of electricity, MW·h	10.7	77172	-	-	Gas-piston station 6xTCG2032V12 Power 3.4 MW, the cost of 57 million rubles
Electricity for the production of oxygen, MW·h	1.3	9360	-	-	
Electricity for own needs of aggregates 1-8, MW·h	0.4	2880	-	-	
Remaining exported electricity, MW·h	10.0	64800	4200	272.2	
Operating costs				60.0	
Profit from sales: el. energy, Al-Si, metal				448.1	
Investments				250	Implementation period, 1.5-2.0 years

*Note: The deliberately inflated value of waste enrichment is accepted, in fact it is equal only to the cost of loading and delivery.

4. The non-volatile scheme of the SER unit

Figure 2 shows one of the variants of a fully energetically closed circuit. The exhaust gas from unit 1 is supplied to reformer 2, which increases the caloric value of the gas to 12320 kJ/m³ and changes its chemical composition to the H₂/CO ratio of 1/1. After gas purification 3, the gas enters the gas piston station 4, on the shaft of which there is an electric generator 5 [7]. As can be seen from Table 4, 7200 m³/h of exhaust gas (after reformer 2) can be fully used in the gas piston station 4 to generate electricity by the generator 5 in the amount of 10.7 MW·h. This makes it possible to completely cover the own needs of aggregates 1-8 (about 0.4 MW·h), produce oxygen in unit 6 (in the amount of 1100 m³/h) and nitrogen (in the amount of 4138 m³/h), with an electricity consumption of about 1.3 MW·h. The remaining part of the electricity in the amount of 9.0 MWh and the thermal energy from the cooling systems of gas piston machines, in the amount of 10.7 Gcal/h, can be used by nearby workshops, process units, micro districts, etc. [8].

A part of the gas, in the amount of 200 m³/h, is pumped by the compressor 7 into the storage gas tank 8 at a pressure of 15 MPa, which acts as a battery. Through a special pressure regulator, the gas enters the gas piston station 4, which makes it possible to ensure its stable operation during short-term idle times of the unit 1.

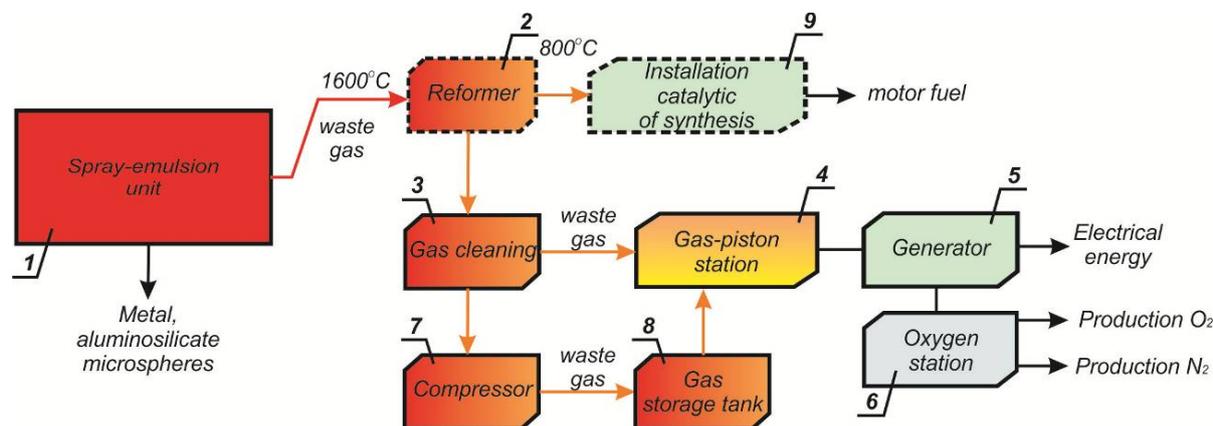


Figure 2. Energotechnological scheme of the installation.

The nitrogen produced at the oxygen station can be used in various production processes, including ammonia synthesis and nitrogen fertilizer production, methane conversion, associated gas processing, as well as for more efficient processing of high-octane components and reduction of sulfur deposits in the production of synthetic motor fuel.

Compared to gas turbines, gas piston engines in the power range up to 4 MW have a higher efficiency and are adapted to work at partial loads [9]. In addition, they are less susceptible to the influence of high ambient temperature, and create less noise and harmful gas emissions [10, 11]. There is already experience in the effective use of reciprocating machines. For example [12], on the territory of the greenhouse complex "UGMK-Agro" an energy complex based on piston machines was built, capable of generating electricity in the volume of 26.44 MW, as well as heat energy, cold and carbon dioxide. The cost of electricity from own generation is 1.9 rubles for kW·h, and from the network 5.4 rubles. This is taking into account the cost of natural gas, on which piston machines operate, and in our case, the gas is obtained from waste coal, for which storage, in principle, you need to pay.

5. Conclusions

Thus, in the presented article it is shown that on the basis of the previously developed spray-emulsion metallurgical SER unit, a completely non-waste technology of coal gasification (processing) and waste enrichment can be created with the production of three liquid products: synthesis gas (or electricity), aluminosilicate micro-granules and iron with rare metals dissolved in it. It should be emphasized that if there are dusty iron-containing waste (dust of gas purification, tailings of enrichment) near the coal enrichment waste, the efficiency of the described technology is increased: the possibility of stable maintenance of the gas-slag emulsion and the flow of the two-phase mixture in the connecting channels is improved, and the consumption of gaseous oxygen due to replacement of oxygen from iron oxides. As can be seen from the table above, the proposed technology has high economic efficiency and quick payback. The implementation of such a technology near the concentrator allows, together with the solution of the problem of processing pulverized waste, to ensure the independence of energy supply.

References

- [1] Tsymbal V P, Mochalov S P, Rybenko I A, Aizatulov R C, Sokolov V V, Padalko A G, Kozhemyachenko V I, Krasnoerov S Yu, Shakirov K M, Kalashnikov S N, Ermakova L A,

- Olennikov A A, Ognev A M, Schipanov S V, Rybushkin A A and Suzdal'tsev E V 2014 *SER Process – Metallurgical Jet-emulsion Reactor* (Moscow: Metallurgizdat) p 488
- [2] Tsymbal V P, Kozhemyachenko V I, Rybenko I A, Padalko A G, Olennikov A A 2015 *Steel in Translation* **45** 550-554
- [3] Terent'ev G A, Tyukov V M, Smal' F V 1989 *Motor Fuel From Alternative Raw Materials* (Moscow: Chemistry) p 271
- [4] Plate N 2004 *Journal Science and Life* **11** 66-68
- [5] Shilling G-D, Bonn B and Kraus U 1986 *Coal Gasification* (Moscow: Nedra) p 175
- [6] Tsymbal V P, Mochalov S P, Rybenko I A Tsymbal Yu V A *Method for Direct Reduction of Metals to Produce Synthesis Gas and an Aggregate For Its Implementation Patent 2371482 Russia C1 priority 03.12.2007 publ. 27.10.2009 Bul* **30**
- [7] Zaychenko V M and Chernyavsky A A 2010 *Academy of Energy* **4 (36)** 58-63
- [8] Olennikov A A and Tsymbal V P 2008 *Steel in Translation* vol 38 **6** 439-443
- [9] Gordeev A V and Mikhailov V A 2017 *International Scientific and Practical Conference* pp 155-159
- [10] Sosnina E N, Masleeva O V, Pachyurin G V and Filatov D A 2013 *Basic Research* **6-1** 76-80
- [11] Rodionova M A, Khrest'yanovskaya M V and Kukolev M I 2017 *Construction of Unique Buildings and Structures* **4(55)** 50-66
- [12] *In UGMK a Unique Energy Center Has Been Built Ore&MetalsWeekly – News of Mining and Metallurgy* <https://www.ugmk.com/press/news/v-ugmk-postroili-unikalnyy-energotsentr>