A NEW SUSTAINABLE PROCESS FOR THE GASIFICATION COAL FINES AND RECOVERING OF METALS AND ALUMINOSILICATES FROM THEIR ASHES

V. Tsymbal¹, I. Rybenko¹, A. Olennikov², P. Sechenov¹, F. Kongoli³

¹ Siberian State Industrial University (Novokuznetsk, Russia);
² Tyumen State University (Tyumen, Russia);
³ FLOGEN Technologies Inc. (Canada);

Keywords: Coal, Gasification, Fines, Metals, Aluminosilicate, SER unit, Gas-slag emulsion, Oxygen conversion, Dimethyl ether, Synthesis gas, Microsphere, Gas-piston power station, Energy & technological scheme

ABSTRACT

This new process makes the gasification of any pulverized coal waste in a suspended layer of slag possible. The process produces a synthetic gas and various metals that are simultaneously reduced from the coal ash, as well as a highly porous aluminosilicate slag-microsphere that can be used in various applications. The input stream consists only of coal from waste while other reactants, including oxygen, are used for burning the coal. These reactants are obtained internally within the same process flowsheet.

This process is based on the previously developed jet-emulsion reactor technology SER originally intended for direct reduction of pulverized metallurgical wastes and ores. It is a sustainable process since it does not produce any waste and is energetically self-sufficient. It can also be designed in a mobile version that can be placed near waste generation sites or in remote places from electrical networks.

INTRODUCTION

In the previous publications [1-4], a new technology developed in cooperation with the specialists of the West Siberian Metallurgical Plant was introduced and described in detail. This new technology is based on a spray-emulsion aggregate SER (self-organizing spray-emulsion reactor) and was initially intended for the direct reduction of pulverized metallurgical wastes and ores. It was shown that this technology has a number of significant advantages over the existing direct reduction processes such as a small specific volume of the reactor unit, high energy intensity and capital intensity [1,2]. These advantages are achieved by using some principles of the self-organization theory, as well as large deviations from thermodynamic equilibrium, large reaction surfaces in the form of gas suspension, forced motion of the working mixture, critical flow of a two-phase medium and other physical principles [1-4]. This technology makes the simultaneous reduction of iron from oxides possible and produces a conditioned synthetic gas within the same reactor [5]. Subsequent studies on the basis of mathematical models have shown the possibility of modifying this process towards the creation of a non-waste technology for gasification of pulverized coal fractions and enriched wastes. This will be the subject of the current paper.

OPERATION PRINCIPLES OF SER UNIT FOR COAL FINES GASIFICATION

The same reactor described in the previous publications [1-4] is used for this new process for the gasification of coal fines. This technology has certain universality because of its possibility to process and/or gasify only coal without the addition of ore or a minimum ore addition in order to reduce the process temperature and produce a foamy slag emulsion. Metals such as iron, manganese etc. are recovered from the oxides contained in the coal ash and in the slag-forming additives while the sum of the CO and H₂ components in the gas approaches 90%. Furthermore, the addition of iron oxides (sludge, tailings of enrichment) to coal is possible and very rational since part of the gaseous oxygen is replaced by oxygen extracted from iron oxides. Additionally, liquid iron and its oxides are catalysts that allow the acceleration of the process and increase the yield of some components in the synthetic gas.

The flowsheet of the process is shown in Figure 1. 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. There, a seal disk is formed at the area where oxygen flows meet opposite counteracting lances 3 which inject the oxygen. The dynamic interaction of the jets creates an intense turbulent stream and, as a result, large surfaces for heterogeneous chemical interactions are formed. Furthermore, due to the incomplete combustion of a part of the coal or of other reducing agents in the reaction chamber 1, heating and partial reduction of oxides occur in accordance with the proportion of oxygen supplied.



Figure 1 – Schematic flowsheet of SER technology for coal fines gasification.

The two-phase gas suspended mixture is naturally pushed through channel 4 in the vertical column refining reactor 5. A significant deviation from thermodynamic equilibrium at this reactor/settler makes possible to separate the metal flow that is deposited in the bottom 7 of the reactor. The flux of the gas-slag emulsion is passed through the inclined channel 11 in a slag receiver 12 where slag and gas are separated.

The produced gas passes into the slag-receiver reactor 12, and then passes through a cooled grate 15 into the gas conversion chamber 14. In order to produce dimethyl ether or methanol, the gas composition is adjusted by constantly replenishing the reactor below the grate 15 with a layer of coke or coal and steam through channel 16.

At the same time, above the grate 15 in the gas reforming chamber 14, natural gas and oxygen are supplied while ensuring that the volume ratio of H_2 to CO in the produced gas is 2/3 or 1/2, respectively, in order to produce dimethyl ether or methanol.

The resulting synthetic gas in the chamber 14 passes through the channel 21 and is fed to a cooler such as waste heat boiler 22, and then to gas purification vessel 23 and finally, to the unit for catalytic synthesis of dimethyl ether or methanol or to the consumer for energy purposes. The sludge generated in the gas scrubbing 23 is recycled into the process which makes the technology waste-free.

An advantage of the present technology is the possibility to control, in a flexible and practically independent way, the composition of the gas at the outlet from the refining settler 5 and at the final outlet of the unit, namely chamber 14. This is made possible by the series nature of the flowsheet containing the refiner 5 and the gas composition reformer 14 combined with the slag receiver 12 and the slag granulator 13.

The combination of the coal gasifier (chamber 14) with the metallurgical units (reaction chamber 1 and refining settler 5) and the slag receiver 12 makes it possible to use the physical heat of associated gas and slag to increase the amount of marketable gas as well as its caloric content. As a result, a chemical regenerator or utilizer of the physical heat is obtained after chamber 14.

The increased pressure created in 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 without the need of forced flow apparatuses.

The gas reformer 14 performs steam-coal combustion and gas enrichment with hydrogen to a volume ratio H_2/CO of 1/1. This is convenient for the subsequent synthesis of dimethyl ether [6,7] in a way that is at least better than in the Fisher-Tropsch method where it is still necessary to separate nitrogen [8].

In contrast, a gas produced in a special gasifier or in a gas generator such as the Lurgi-Fischer-Tropsch technology requires a considerable amount of thermal energy to maintain the temperature in the gasification chamber at 800-1000°C and for nitrogen heating [8]. Furthermore, in the Lurgi-Fischer-Tropsch technology the gas obtained has low caloric value and requires large expenditure for the separation of ballast components before catalytic synthesis.

PRODUCT CHARACTERISTICS

This technology can produce highly porous aluminosilicate microspheres due to the fact that the gas suspension in the column reactor 5 has a high gas content of more than 99%. The gas suspension passes at high speed through the incline connecting channel 11 to the slag receiver

vessel 12. 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 motion regime of the two-phase working mixture prepared in the reactor-oscillator (the first reactor) is achieved. This way, the conditions are fulfilled when the slag film is attached on gas bubbles and hollow spherical structures (microspheres) are formed. By managing the slag regime, it is possible to obtain films of the required chemical composition. The installation of a gravity separator after the slag-receiver granulator makes possible the classification of the microspheres according to the granulometric composition by satisfying the most stringent requirements. The micro-granules of high quality are more valuable in the market compared to metal. The chemical composition of slag micro-granules obtained through the mathematical model for the gasification of coal waste is given in Table 1.

Table 1. –	Chemical	composition	of micro	ogranules.
1 4010 1.	Chenneur	composition	or more	Si alla los.

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

After the steam conversion, chamber 14 produces a gas whose composition is given in Table 2. The H_2/CO ratio of 1/1 makes it suitable for the catalytic synthesis of motor fuels.

T 11 0	C '.'	C C		•
Table 2. $-$	Composition	n of gas affer	steam-oxygen	conversion.
10010 -	0011100011101			•••••••••••••••••••••••••••••••••••••••

Gas	CO	CO ₂	N ₂	H ₂	H ₂ O	CH4	Total
Wt %	51.518%	31.355%	0.887%	3.680%	12.559%	0.000%	100.00%
Volume %	35.92%	13.91%	0.62%	35.92%	13.62%	0.00%	100.00%

Alternatively, this gas can be fed to a gas turbine or a gas piston machine to produce electricity. This completes the first stage of the project's implementation.

If the task is to convert the synthetic gas into motor fuel, the abovementioned gas can be used well. Furthermore, by using oxygen conversion, the amount of methane can be 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 5 Composition of gas aren additional methane oxygen correction.							
Gas	СО	CO ₂	N_2	H_2	H ₂ O	CH4	Total
Wt %	58.084%	29.434%	0.880%	5.632%	5.970%	0.000%	100.00%
Volume %	35.03%	11.30%	0.53%	47.55%	5.60%	0.00%	100.00%

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

The metal (iron-carbon melt) that is tapped from tapping hole 8 helps maintain a stable gas-slag emulsion where carbon and volatile constituents of coal are burned. In addition, iron oxide is also a catalyst for the formation of hydrocarbons. The metal, periodically produced from the fore hearth, can simply be sold as a first-class scrap whose market is unlimited. Since, in principle, all metals contained in the coal ash are concentrated in this metal, achieving this task task requires a separate study.

In the presence of a conditioned synthetic gas, the task of catalytic synthesis is not difficult, although, of course, it requires additional, but quickly recouped, capital investments. This possible development is not considered here.

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

Input current	Consump /hour	Consump /year	The price of units, USD/ton, or thousand m ³	Costs, income per year, USD	Notes			
Coal (waste), ton	3.6	25920	6.24*	162,292.00				
Oxygen, m ³	1075	322500	-	-	Oxygen station (with compressor up to P=150 atm.) – 124,880.00 USD			
Products								
Gas, thousand m ³	7.2	51840	62.42	3,245,319.00				
Al – Si, tons	0.9	6480	546.20	3,540,348.00				
Metal, tons	0.07	504	280.89	142,051.00				
Possible full generation of electricity, MWh	10.7	77172	-	-	Gas-piston station 6xTCG2032V12 Power 3.4 MW, the cost of 889,533.91 USD			
Electricity for the production of oxygen, MWh	1.3	9360	-	-				
Electricity need in reactors 1 to 8, MWh	0.4	2880	-	-				
Remaining exported electricity, MWh	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 this value is equal only to the cost of loading and delivery.								

Table 4. – Technical and economic indicators.

The efficiency 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 a profit of 265.28 USD. It should be emphasized this is a completely ecologically and energetically closed technological flowsheet whose input is only coal (waste) and products include alumino-silicate microgranule, metals, gas as well as oxygen. Another advantage is that this makes it possible to design this technology unit as a mobile version and place it near waste generation sites or in places remote from electrical networks.

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 goes to reformer 2, in which the caloric value of the gas rises to 12320 kJ/m³, and its chemical composition changes to a ratio of H₂/CO equal to 1/1. After the gas cleaning 3, the gas enters the gas piston station 4, on the shaft of which is electric generator 5 [9].



Figure 2 – Technological flowsheet of the installation.

As can be seen from the data given in the Table 4, 7200 m^3 /h of exhaust gas can be used after reformer unit 2. This makes it possible to build up an oxygen unit (unit 6) with a capacity of 1100 m^3 /h and a nitrogen unit with a capacity of 4138 m^3 /h with an electricity consumption of about 1.3 MWh. The remaining part of the electricity of about 9.0 MWh and the thermal energy from the cooling systems of gas piston machines of about 10.7 GCal/h can be used by nearby workshops, process units, micro-districts, etc.

A part of the gas of about 200 m³/h is pumped by compressor 7 at a pressure of 15 MPa into the storage gas tank 8, which acts as a battery. Through a special pressure regulator, the gas enters the unit 4, which ensures their stable operation with short-term outages of unit 1.

CONCLUSION

In the presented article, it is shown that on the basis of the previously developed spray-emulsion metallurgical SER unit, a complete non-waste technology for the coal gasification and waste enrichment was developed. It provides three liquid products: synthetic gas (or electricity), aluminosilicate micro-granules and iron with rare metals dissolved in it. The efficiency of the technology increases if dusty iron-containing waste (dust of gas purification, enrichment tailings etc.) is used from the waste coal enrichment sites. This is because this creates the possibility to achieve a much more stable gas-slag emulsion and improves the flow of the two-phase mixture in the connecting channels, as well as reduces the consumption of oxygen by replacing it with the oxygen coming from iron oxides. The proposed technology has high economic efficiency and fast payback. Furthermore, its implementation near industrial plants solves not only the problem of processing of pulverized waste but also ensures the independence of the energy supply.

REFERENCES

[1] V.P. Tsymbal, S.P. Mochalov, I.A. Rybenko, R.S. Ayzatulov, V.V. Sokolov, A.G Padalko, V.I. Kozhemyachenko, S.Y. Krasnopyorov, K.M. Shakirov, S.N. Kalashnikov, L.A. Ermakova,

A.A. Olennikov, A.M. Ognev, S.V. Szczepanov, A.A. Rybushkin, E.V. Suzdaltsev, Moscow: Metallurgizdat, (2014) 488.

[2] Tsymbal V, Olennikov A, Kozhemyachenko V, Rybenko I, Kongoli F. 2015. Efficient Utilization Of Fuel Energy In The Steel Industry Using A New Structure Of Energy And Metallurgical Unit Type Self-Organizing Spray-Emulsion Reactor. In: Kongoli F, Kleinschmidt G, Pook H, Ohno K, Wu K, editors. Sustainable Industrial Processing Summit SIPS 2015 Volume 2: Gudenau Intl. Symp. / Iron and Steel Making. Volume 2. Montreal (Canada): FLOGEN Star Outreach. p. 259-266.

[3] Tsymbal V, Olennikov A, Rybenko I, Kozhemyachenko V, Protopopov E, Kongoli F. 2016. Basic Principles And Features Of Self-Organizing Jet-Emulsion Technology (SER). In: Kongoli F, Noldin JH, Takano C, Lins F, Gomez Marroquin MC, Contrucci M, editors. Sustainable Industrial Processing Summit SIPS 2016 Volume 1: D'Abreu Intl. Symp. / Iron and Steel Making. Volume 1. Montreal(Canada): FLOGEN Star Outreach. p. 214-227.

[4] Tsymbal V, Olennikov A, Rybenko I, Sechenov P, Kongoli F. 2017. Mathematical Modeling of SER Jet-Emulsion Process. In: Kongoli F, Conejo A, Gomez-Marroquin MC, editors. Sustainable Industrial Processing Summit SIPS 2017 Volume 9: Iron and Steel, Metals and Alloys. Volume 9. Montreal (Canada): FLOGEN Star Outreach. p. 104-115.

[5] V.P. Tsymbal, S.P. Mochalov, I.A. Rybenko, Yu.V. Tsymbal, Patent 2371482 Russia C1, Publ. 27.10.2009, Bul. № 30.

[6] G.A. Terent'ev, V.M. Tyukov, F.V. Smal', Moscow, Chemistry (1989) 271.

[7] N. Plate, Journal Science and life (2004) № 11, 66-68.

[8] G-D. Shilling, B. Bonn, U. Kraus, Moscow, Nedra (1986) 175.

[9] A.V. Gordeev, V.A. Mikhailov, International Scientific and Practical Conference (2017), 155-159.