Investigation for a Sustainable Use of Fossil Coal through the Dynamics of Interaction of Smokeless Solid Fuel with Oxygen

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Abstract

One of the ways to solve the problem of environmental protection in the fuel/metallurgical sectors of the economy is the introduction of new complex technological processes, which can be implemented by the principle of energy-chemical-technological processing of carbon raw materials. It involves the extraction of valuable chemical components in the form of coke oven gas, benzene hydrocarbons, hydrogen sulphide, coal tar, ammonia, as well as the production of solid product with new properties.

A technology for the production of smokeless solid fuel from a mixture of gas coal (80%) and coal wastes (20%) has been developed. Comparative studies of the kinetics of combustion have been carried; the dependence of the rate of interaction of coke residues of anthracite, highly metamorphosed coal, low-metamorphosed coal and smokeless solid fuel with oxygen of air was obtained. The characteristics of anthracite, high-metamorphosed coal, low-metamorphosed coal and smokeless solid fuel combustion dynamics are obtained, which allow further determination of the reactivity of the carbonaceous substance of the fuel.

The developed technology of production of smokeless carbon fuel offers the opportunity to use carbon wastes and improve environmental performance. According to the results of industrial experiments, the optimal quality indicators of smokeless solid fuels recommended for ferroalloy industries were established at ferroalloy enterprises.

Keywords
Coal, Smokeless solid fuel, Combustion kinetics, Combustion dynamics, Ferroalloys

1. Introduction

One of the ways to solve the problem of resource conservation and environmental protection in the fuel and energy and metallurgical sectors of the economy is the introduction of new complex technological processes. To solve this problem, Zbykovsyy (2019, 2018), Golubev et al. (2017a, 2017b) and Zolotaryov et al. (2011) developed the new technology based on the principle of energy-chemical-technological processing of carbon raw materials. It involves the extraction of valuable chemical components and compounds in the form of coke oven gas

(combustion heat $\sim$ 17700 kJ/m^3), benzene hydrocarbons, hydrogen sulphide, coal tar, ammonia and other chemical compounds, as well as the production of solid product with new consumer properties.

Aronov et al. (1968) and Korchevoi et al. (2004) found that smokeless solid fuel in its technological and technical characteristics can be compared with anthracites. In the future, it may serve as an alternative to their use. At the same time, this fuel can be an alternative to coke nuts in ferroalloy production due to its high reactivity.

Smokeless solid fuel is a product of high-temperature coking of a pre-heated coal charge, which consists of low-metamorphosed low-sulfur coal (80-100%) and coal-waste (up to 20%). The technology for producing smokeless solid energy fuel was developed by Zbykovskyy et al. (2008, 2009).

The main technological characteristics of smokeless solid fuel are:
- Total Sulphur content of S_t^d < 1%;
- Yield of volatile substances V^{daf} = 1-3%;
- Ash content A^d = 10-20%;
- Lower combustion heat $\geq$ 26000 kJ / kg;
- Reactivity CRI$\geq$65%;
- Maximum heat dispersion rate at T> 860^{0}C.

The technology of obtaining carbon smokeless solid energy fuel was developed in 3 stages by Gordienko et al. (2009, 2008, 2007). There are three main stages: preparation of the initial coal mixture (1st stage), thermal preparation of coal mixture (2nd stage) and coking of preheated mixture (3rd stage).

2. Methodology and research approach

To study the kinetics of the interaction of the carbonaceous substance of the fuel with the gas-reagents at a pressure of 0.1 MPa and at a temperature up to 1200^{0}C, a laboratory unit was used, which a differential impulse reactor is operating in modes close to perfect extrusion by the gas phase and perfect mixing solid phase. This laboratory unit also allows maintaining small temperature gradients (less than 50^{0}C) and pressures (less than 100 Pa) in the reaction zone over the height of the fuel layer when conducting experiments.

Intense mixing of the fuel particles in the fluidized zone provides a mode of perfect mixing of the solid phase, as well as almost homogeneous concentration of the gas-reagent over the layer of analysed solid fuel. The current nature of the movement of gas in the reactor provides a mode of perfect extrusion by the pulse of the gas-reagent.

The experiments and processing of the obtained data were carried out in accordance with a specially developed methodology. The order of measurements determined by the purpose of the kinetic experiment:
- Study of the dependence of the reaction rate on the conversion level at a given temperature (R_m = f (X) at T = const);
- Study of the dependence of reactivity rate from temperature (R_m = f (T)).

To obtain the dependence of the burning rate on the conversion level of the fuel sample, the experiment was conducted as follows. A pre-dried and weighed sample of fuel weighing 0.1g was placed on a reactor's perforated grate, mounted in a cold furnace. The reactor was then purged with inert gas (argon or helium) while heating the furnace at a speed of 8-10^{0}C/min to the desired temperature. After temperature stabilization, the impulses of the gas-reagent were sent to the reactor. Under impulse conditions, a predetermined portion of the gas-reagent from the calibrated

loop was introduced into the stream of inert gas by a six-way valve, which was continuously inserted into the lower part of the reactor. During the passage of the impulse, the interaction of the gas-reagent with the fuel sample and the formation of reaction products occurs. The component composition of the reaction products is determined at the reactor's output by the gas-analyser. Thus, the experiment involves the sequential introduction of impulses of the gas-reagent and the measurement of the formed products' concentrations.

The duration of the impulse response at the output of the reactor is measured by a chronometer according to the measurements of the rheometer / rotometer. The jump-alike change of density during the transition from inert gas to the gas-reagent and back leads to a jump-alike change in the measurements of the rheometer, which allows determining the moment of occurrence of the impulse-response. The composition of combustion products (CO, CO_2, O_2) is measured by the gas-analyser.

Experiments to determine the dependence of the reaction rate on temperature were also carried out using impulse supply of gas-reagent. The sample of solid fuel is placed in the reactor. Thereafter, the reactor continuously heated to a predetermined temperature with simultaneous purging of the inert gas to prevent the fuel sample from interacting with the oxygen of the air. The component composition of the gaseous products is determined by the gas-analyser at the output of the reactor.

For a given temperature level, the impulse input lasts no more than one or two times depending on the sample conversion rate. After that, it goes to a higher temperature level (the temperature step is $25\text{-}50^0C$). The stepwise temperature increase, followed by the analysis of the gaseous products of the impulse response, carried out until the maximum required temperature is reached.

In all experiments, the impulse value of the gas-reagent is chosen in the way that the mass reduction of the solid fuel sample (Δm) during the impulse (Δt) would be much less than its initial value (m_0) and also based on the limiting sensitivity of the used gas-analyser. During the experiments, the impulse value of the gas-reagent, given by the volume of the loop, is tucked up experimentally.

Samples of coal used for research: anthracite, highly metamorphosed coal, low-metamorphosed coal, and smokeless solid fuel (Table 1). For kinetic studies, coke residues of the presented solid fuels were prepared at a temperature of 850^0C for 30 minutes in an inert gas stream. Working with coke residues of fuel during kinetic studies is a prerequisite, since it allows determining the reactivity of coke fuel residue, which is responsible for the longest stage of combustion of fuel particles, without the specific properties of volatile substances.

Table 1. Technical and elemental analysis of the researched types of solid fuel

Type of solid fuel	Technical analysis			
	W^a, %	V^{daf}, %	A^d, %	S^d_t, %
Anthracite	2.3	2.00	4.39	0.75
Highly-metamorphic coal	1.8	11.10	10.90	1.54
Low-metamorphic coal	3.3	41.60	6.20	0.46
Solid smokeless fuel	1.9	2.70	11.80	0.74
Type of solid fuel	Elemental analysis			
	C^{daf}, %	H^{daf}, %	N^{daf}, %	O^{daf}, %
Anthracite	96.27	2.20	0.92	0.61
Highly-metamorphic coal	91.00	3.98	2.28	2.74
Low-metamorphic coal	81.62	5.95	1.61	10.82
Solid smokeless fuel	97.89	0.68	0.73	0.70

3. Results and Discussions

As a result of kinetic experiments, the dependence of the interaction rate of anthracite coke residue with oxygen at a constant temperature $T = 560^0C$ was determined. The dependence of the anthracite sample burnout rate (R_{m0}), normalized to the initial mass of fixed carbon in the sample, and is presented (Figure 1a). Kinetic experiments performed on samples weighing about 0.1g with fuel particles with a diameter of 0.1-0.16mm.

The total flow rate of the gas-reagent through the reactor was 1.2-1.6 dm^3/min at atmospheric pressure. The speed of burnout is extreme and varies with the conversion of the sample: increases to the maximum value ($R_{m0} \cong 4\cdot10^{-4}$, 1/s at X = 25-40%) and decreases in the final stage of burnout. The maximum conversion level of the sample of anthracite coke residue at a temperature $T = 560^0C$ was 80-82%.

The dependence of the rate of interaction between the lean coal coke residue and oxygen on the conversion level ($T = 560^0C$, d = 0.1-0.16mm) was also investigated (Figure 1b). As with anthracite, the dependence of the rate of burnout is non-stationary during the conversion: passing through the maximum value ($R_{m0} \cong 7.4\cdot10^{-4}$, 1/s) at the very beginning of the conversion curve (X = 3-5%), the speed decreases monotonically at the final stage of burnout.

The burnout rate of coke residue of highly metamorphosed coal at a given temperature over the entire range of conversion level is higher than the rate of burnout of anthracite coke residue. The maximum conversion level of the coke residue sample of highly metamorphosed coal at a temperature $T = 560^0C$ was 90-92%.

During the kinetic experiments, the dependence of the interaction rate of the coke residue of low-metamorphosed coal with oxygen at a constant temperature $T = 560^0C$ was established (Figure 1c). Kinetic experiments performed on samples weighing 0.1g for particles' diameter d = 0.1-0.16mm, while maintaining other parameters equal.

As in the previous cases, the dependence of the burnout rate changes during the conversion of the sample: passes through the maximum value ($R_{m0} \cong 4.8\cdot10^{-3}$, 1/s) in the region of the conversion level (X = 20%) and decreases at its final stage. The burnout rate of coke residue of low-metamorphosed coal is several times higher than the burnout rate of both high-metamorphosed coal and anthracite in the entire range of conversion level and is characterised by the highest reactivity (burnout rate). The maximum conversion level of coking coal residue at temperature $T = 560^0C$ was 92%.

The dependence of the interaction rate of the coke-residue of smokeless solid fuel with oxygen with the conversion level ($T = 560^0C$, d = 0.1-0.16mm) was also investigated (Figure 1d). As with the previous types of fuel, the burnout rate depends on the conversion level of the sample: passes through the maximum value ($R_{m0} \cong 4.2\cdot10^{-4}$, 1/s) at the very beginning of the conversion curve (X = 6-8%), and then monotonically decreases in the final stage of burnout. The smokeless coke residue rate at this temperature ($T = 560^0C$) at the initial conversion site exceeds the anthracite burnout rate. At the same time, it decreases during the conversion below the burnout rate of low-metamorphosed and highly metamorphosed coal.

The maximum conversion level of coke-residue of smokeless solid fuel at a temperature of $T = 560^0C$ was 33-35%, which can be explained by the significant deactivation of the active surface of coal during the previous high-temperature carbonization.
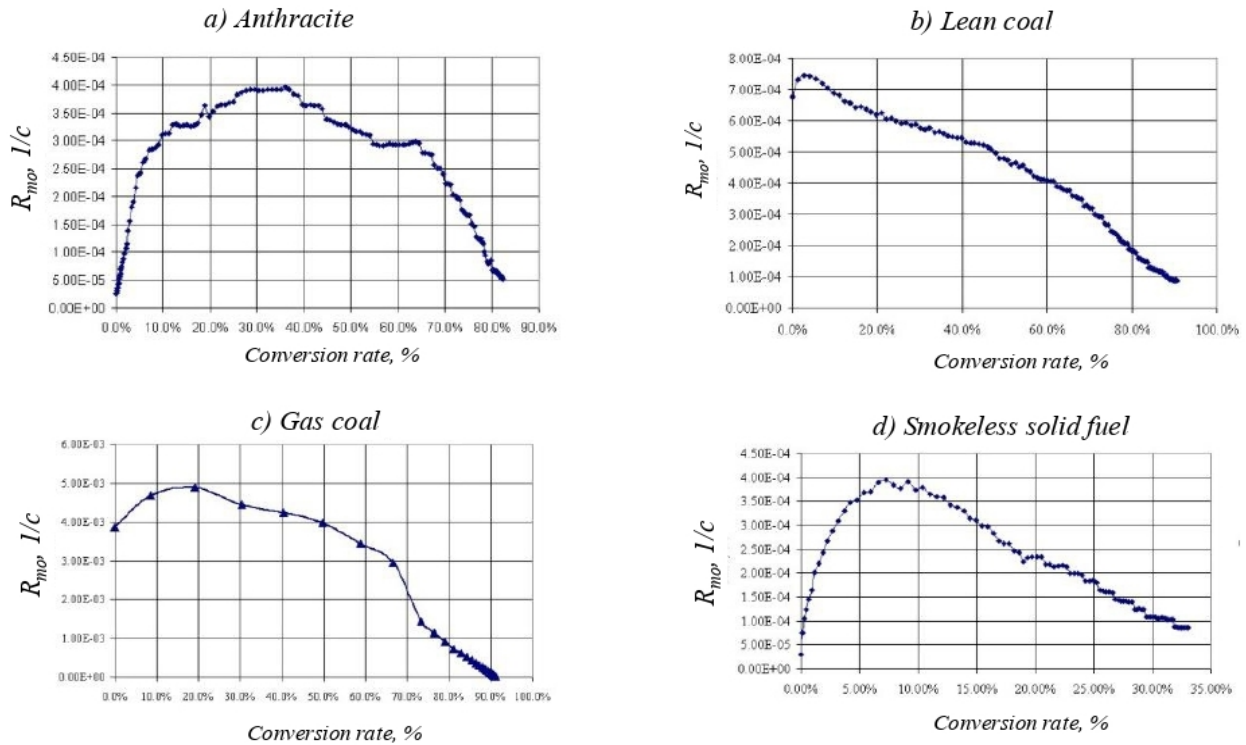
Figure 1. Interaction rate for various types of energy fuel with oxygen and conversion degree

The summary graphics of the results of the experiments allow us to compare the obtained dependences of the speed of burnout of different types of solid fuels on the conversion level (Figure 2, Figure 3). In the entire range of conversion, the burnout rate of coke-residue of anthracite particles is the lowest compared to the burnout rate of high-metamorphosed and low-metamorphosed coal. The dependence of the burnout rate of smokeless solid fuel at a given temperature is intermediate and characterized by low reactivity, which is the result of deactivation of the activated carbon surface in the process of preliminar high-temperature heating without air access (Vilenskii et al., 1978; Pomerantsev, et al., 1986).
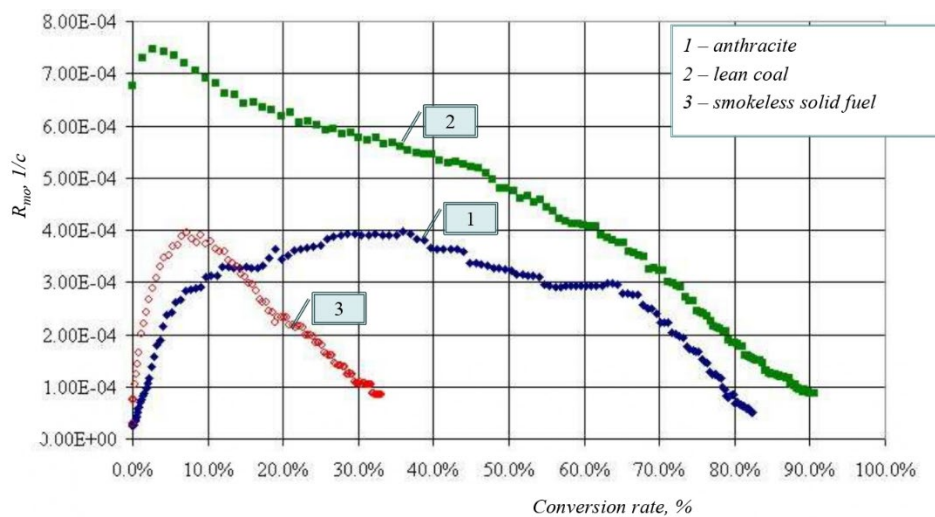

Figure 2. The relationship of the interaction rate of energy fuel with oxygen and conversion degree
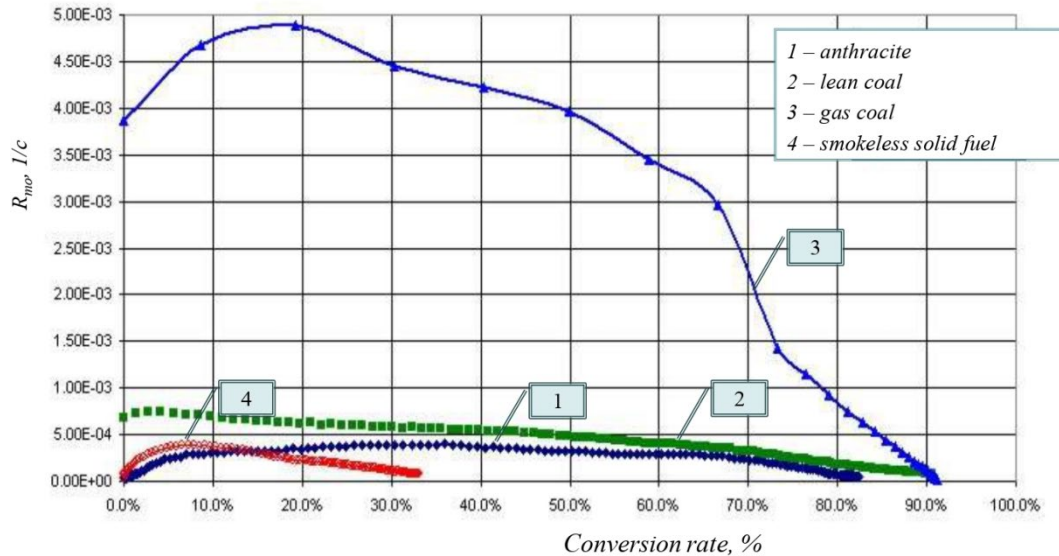
Figure 3. Interaction rate of energy fuel with oxygen and conversion degree compared to gas coal

In the experimental studies of the burnout dynamics of anthracite, high-metamorphosed, low-metamorphosed coal and smokeless solid fuel, the samples of the original fuels' particles were used. Experiments of burnout of the carbon particles performed at starting temperatures close to 860^0C. In order to ensure an accurate comparison of results, very identical experimental conditions were maintained for all coal samples (initial temperatures, sample masses, air flow rates, particle sizes, etc.). The registration of the temperature measurements in the reactor was stopped after reaching the minimum values of concentration (limit of measurement) of the reaction products (CO and CO_2) according to the gas-analyser (for CO – 0.001% vol., for CO_2 – 0.01% vol.). Thus, the burnout time corresponds to the full burnout time of the coal particles and is determined equally for all samples of the tested coals. Experimental studies on the dynamics of burnout modelling the natural conditions of particles' burnout in a furnace/boiler.

The dynamics of the temperature change in the reactor during the burnout of discrete anthracite samples with diameter d = 0.1-0.16mm at atmospheric air pressure was investigated (Figure 4a). The weight of the sample in all experiments was maintained constant ($\cong$1 g) with a coal fraction of 0.3g and ash at 0.7g, taking into account the constancy of carbon in all samples. The linear speed of the gas stream was calculated taking into account the prevention of removal from the reactor of fuel particles of the minimum size (Ug = 0.38 m/s, T = 900^0C, P = 0.1 MPa).

During the burnout of the anthracite sample, a dramatic change detected in the character of the heat release in the reactor and temperature - from initial (about 860^0C) to 945-950^{0}C. The temperature curve of the anthracite burning corresponds to the burning of carbon of the coke-residue, and does not contain the initial area of the release and burnout of volatile substances. The effective burnout time of anthracite particles for a given high temperature (860^0C), corresponds to values of about 490 sec. and is the highest value of all the types of studied fuels.

A similar burning curve was obtained for particles of high-metamorphosed coal, all experimental parameters were maintained equal (Figure 4b). Its peculiarity is the existence of two extreme areas: the first extreme area corresponds to the burnout of volatile substances over a small interval of time; the second extreme area corresponds to the burning of the coke-residue over a longer time interval. The burnout duration for particles of high-metamorphosed coal for a given high temperatures (860^0C) was about 425 seconds.

The dynamics of burnout of discrete samples of low-metamorphosed coal is characterized by the presence of two distinctive peaks: the first peak is short and corresponds to burnout of volatile substances (Figure 4c); the second peak is more durable, and corresponds to burnout of coke-residue. The burnout duration for particles of low-metamorphosed coal for a given high temperatures (860^0C) was the smallest – about 280 seconds.

Although the smokeless solid fuel is derived from low-metamorphosed coal (with content of volatile matter V^{daf} = 41.6% vol.), the temperature curve of the discrete smokeless solid fuel burnout does not contain a peak corresponding to the yield and burnout of volatile matter. The temperature curve corresponds to burnout of the coke-residue (Figure 4d). The burnout duration of the smokeless solid fuel particles for a given high temperature (860^0C) was about 400 seconds.
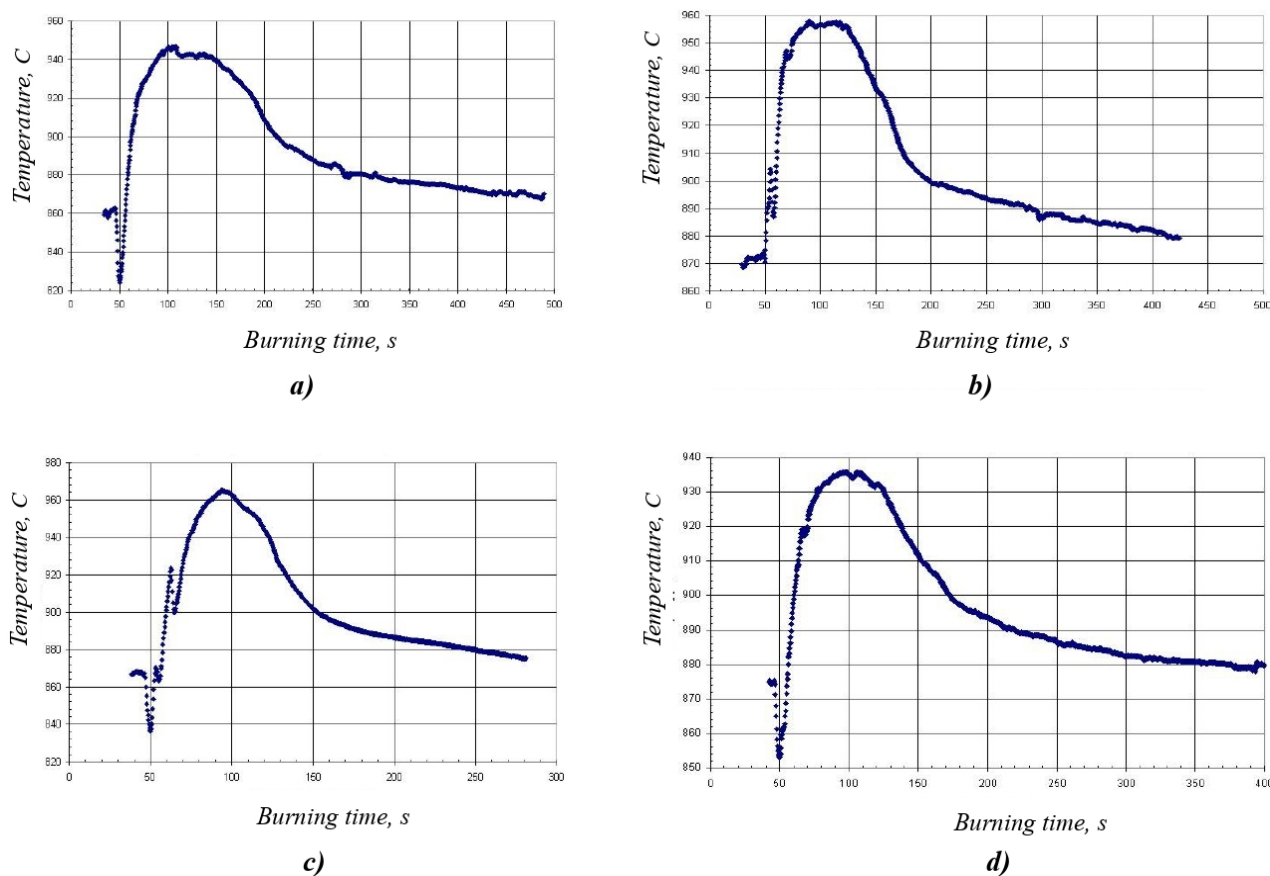

Figure 4. Dynamics of combustion of coal particles of anthracite (a), lean coal (b), gas coal (c) and smokeless solid fuel (d) by reaction with oxygen and atmospheric pressure

Comparative characteristics of dynamic curves of burnout of the studied fuels (Figure 5) show that the shortest burnout duration has a low-metamorphosed coal, then - smokeless solid fuel, high-metamorphosed coal, and the highest – anthracite. The obtained temperature curves allow us to analyse the level of maximum heat release during the burnout of the investigated fuels, the duration of the pyrolysis stages and the burnout of coke-residues, the complete burnout time and the reactivity of these solid fuels (Vilenskii et al., 1978; Pomerantsev et al., 1986).
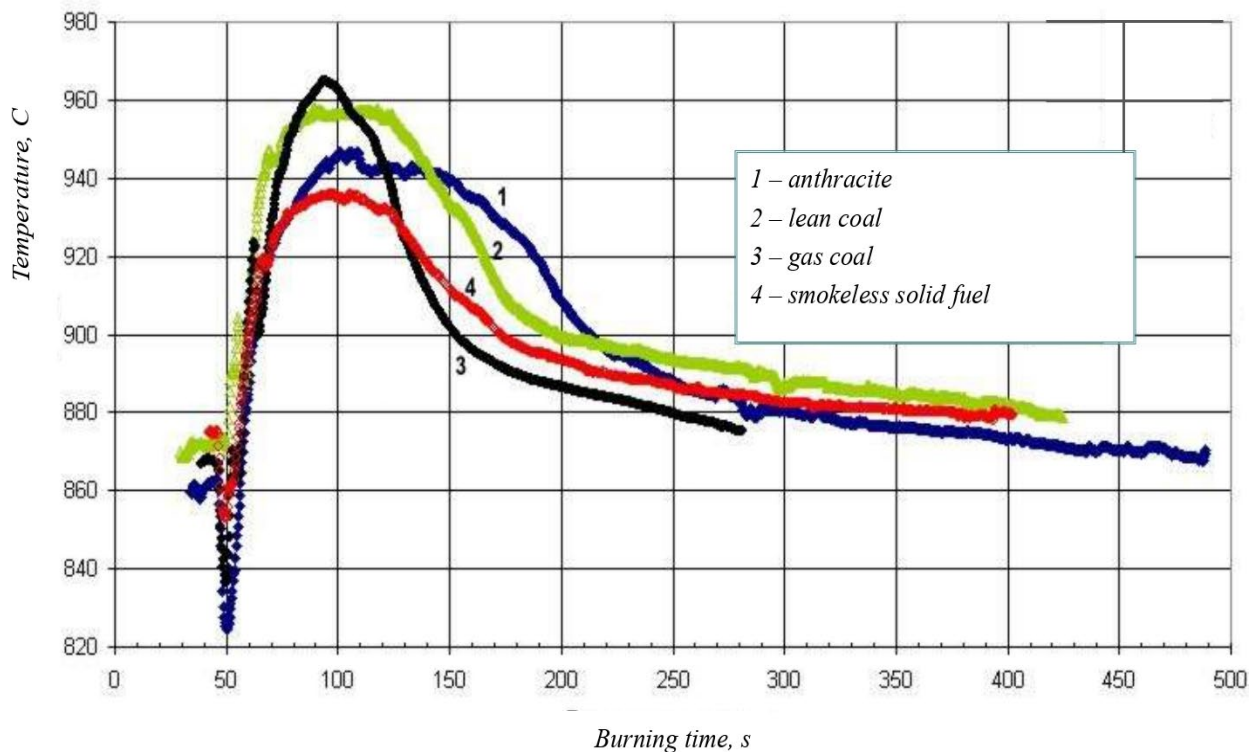
Figure 5. Dynamics of energy fuel combustion

Yield and burnout of volatile matters of the fuel contributes to its ignition and further burnout when get to the furnace. The discrepancy of the characteristics between the investigated fuel and the projected one, in particular, the absence of volatile matters, can lead to an increase of the induction time, its late ignition and, as a consequence, an increase of the mechanical underburn, which will lead to a deterioration of the technical and economic performance of the power-generation unit (Korchevoi et al., 2004).

The possible use of solid carbon fuel is the production of ferroalloys in electric furnaces. To test the efficiency of the use of smokeless solid fuel, its experimental samples were elaborated on the coke battery № 3 of the Yasiniv Coke Plant (Ukraine). During the production of smokeless solid fuel according to the proposed technology, were used three variants of the composition of the charge, which consisted of low-metamorphosed coal, bituminous coal ($W^r = 9.0\%$; $A^d = 8.5\%$; $S_t^d = 1\%$; $V^{daf} = 32.0\%$; $Y = 18$ mm) and coal wastes ($W^r = 12.0\%$; $A^d = 10.0\%$; $S_t^d = 1.2\%$; $V^{daf} = 31.0\%$; $Y = 10$ mm). An experimental sample of smokeless solid fuel used in the experimental smelts for the production of ferro-silicon manganese at furnace № 8 of the Nikopolsky Ferroalloy Plant and at furnaces № 1 and № 2 of the Stakhanovsky Ferroalloy Plant (Table 2).

As shown by the experimental smelts at the Nikopolsky Ferroalloy Plant, the use of highly reactive smokeless solid fuel did not lead to changes in the gas and electrical modes of ferro-silicon manganese smelting (Table 3). There was a lower fit of the electrodes into the charge during the experimental period by 150-200 mm, which indicates a greater adaptability of the use of smokeless solid fuel.

Table 2. The quality indices of the charge and solid smokeless fuel

Consumers	Charge composition, %			Technical analysis of the charge,%			Technical analysis of the solid smokeless fuel, %		Screen composition of the solid smokeless fuel, %				CRI, %	CSR, %
	G	Coal processing waste	Zh	A^d	S^d	V^d	A^d	S^d	>40 mm	25-40 mm	10-25 mm	0-10 mm		
Nikopolsky ferro-alloys plant	80	10	10	8.2	0.8	40.3	11.7	1.2	4.0	8.0	77.0	6.0	65	18
Stakhanovsky ferro-alloys plant	80	10	10	8.0	0.75	39.4	11.5	1.1	2	12	78.0	8.0	61	21
Stakhanovsky ferro-alloys plant	100	0	0	7.2	0.5	41.5	10.5	0.7	2	10	78.0	10.0	76	9

During the experiment, the weight of coke decreased from 360-370 kg to 320-330 kg. The quality of ferro-silicon manganese obtained during the experiment, the content of manganese (72-73%), silicon (17.6-17.8%) and phosphorus (0.25-0.28%) did not differ from the standard values; the specific consumption of manganese raw materials, coke and quartzite decreased respectively by 1.1; 16.6 and 4.4 kg/t, and electricity decrease – by 41.9 kWh/t. The content of manganese in the slag decreased by 1.2%, the extraction of manganese increased by 0.7%, the productivity of the furnace increased by 1.2% (Table 3).

Table 3. The melting indices of the MnC17 grade ferro-silicon manganese
in furnace № 8 of the Nikopolsky ferro-alloys plant using highly reactive solid smokeless fuel

Indices	Melting process options	
	Standard	Pilot
Unit consumption of raw materials, kg/t:		
− manganese raw materials (48% Mn)	1667.4	1666.3
− ordinary coke	440.9	–
− solid smokeless fuel	–	424.3
− quartzite	393.9	389.5
− iron ore pellets	37.4	29.9
− secondary manganese raw materials	247.7	244.5
− waste	367.4	188.3
− screenings	60	62.3
Unit consumption of electric power, kW·h/t	4331.4	4289.5
Manganese to alloy transition, %	86.6	87.3
Manganese content in slag, %	12.3	11.5

During the experiment in the conditions of the Stakhanovsky Ferroalloy Plant, the specific consumption of quartzite and reducing agents – in terms of carbon – decreased by 0.4% and 1.06%, respectively, and the electricity consumption decreased by 375 kWh/t (Table 4). As a result of the industrial experiments, the optimum quality indicators of smokeless solid fuels recommended for ferroalloys were established at the Nikopolsky Ferroalloy Plant and the Stakhanovsky Ferroalloy Plant (Table 5).

Table 4. The melting indices of the FS65 grade ferrosilicon using highly reactive solid smokeless fuel

Indices	Melting process options	
	Standard	**Pilot**
Working hours:		
– rated, days	52.5	26
– hot downtime, %	0.6	0.14
– actual, days	52.21	25.86
Production FS 65, t	3578.8	1881.14
The average silicon content, %	66.3	66.5
Active power, kW	20960	21146
Productivity, t/days	68.2	72.7
Unit consumption, t/t: - quartzite	1.517	1.511
- ordinary coke	0.620	–
- solid smokeless fuel	–	0.609
- gas carbon	0.148	0.116
- carbon reducer	0.5829	0.5567
- steel cuttings	0.340	0.343
- scrap metal of the ferrosilicon	0.058	0.048
- limestone	0.014	0.115
Electric power consumption, kW·h/t	7338	6963
Silicon extraction, %	92.1	92.5

Depending on the brand of alloys and the type of new reducing agents, the proposed technology allows:

- to increase the efficiency of ferroalloy furnaces of the by 5–10%
- proportional reduction in the specific energy consumption by 5–7%
- to increase the proportion of the main elements that pass into the ferroalloys.

A study of the properties of smokeless solid fuels has shown that this fuel meets the requirements for reducing agents for ferroalloys' production plants.

Table 5. Recommended quality indices of the solid smokeless fuel for ferro-alloys production

Quality indices	Symbols	Value
Technical analysis:		
ash content, %	A^d	23.0
total sulphur content, %	S_t^d	1.15
light constituents yield, %	V^{daf}	1.9
Size distribution: +80 – 32.7%; 60-80 – 30.0%; 40-60 – 23.6%; 25-40 – 6.7%; <25 – 7.0%		
crushing strength of coke, %	M_{25}	81.3
coke abrasion resistance, %	M_{10}	13.1
Apparent density, g/cm^3	d_k	0.935
True density, g/cm^3	d_r	1.9301
Porosity, %	p^d_r	51.6
Reactive capacity according to DSTU 7664:2014, $cm^3/g \cdot c$	K_m	1.67
Coke reactivity according to ISO 18894, %	CRI / CSR	69.8 / 30.1

According to the results of performed laboratory, experimental and pilot studies of smokeless solid fuel, the requirements for its quality indicators have been developed for its usage in gasification technologies, production of sugar, soda, cement, during limestone burning/calcination.

4. Conclusions

The technology of production of smokeless carbon fuel has been developed during this research work. The result of the processing is lumpy carbon fuel, which has a homogeneous solid structure, high combustion and mechanical strength, high reactivity, high speed and complete combustion of the fuel particles. Proposed technology offers the opportunity to **reutilise carbon wastes** and **improve environmental indicators** of the air pollution.

Smokeless lumpy carbon fuel has several advantages over other types of fuel: combustion of fuel particles in high-temperature zones – more than 800^0C – transfer combustion mode to a phase close to exterior diffusion. Based on the obtained experimental/industrial results, the **optimum quality indicators** of smokeless solid fuels recommended for ferroalloy industries were established and successfully tasted.

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