Research Article

Formation of Liquid Products at the Filtration Combustion of Solid Fuels

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Yields of liquid and gaseous products of the filtration combustion of cellulose, wood, peat, coal, and rubber have been investigated. Experiments have shown that the gasification of solid fuels in the regime with superadiabatic heating yields liquid hydrocarbons with quantity and quality, which are close to those produced using other methods, for example, by pyrolysis. But in this case no additional energy supply is needed to carry out the gasification process. The low calorific combustible gas, which forms in this process, contains a substantial quantity of carbon monoxide and hydrogen, which are components of syngas.

1. Introduction

Major advantages of the fossil solid fuels (such as oil shale, brown coal, and peat) are their relative abundance and availability of deposits in developed regions and often a previously underused built mining industry. However, these fuels have drawbacks, which hamper their extensive using in power generation. The first is low efficiency of the Rankin steam cycle. The disadvantages of low energy efficiency, problems with fly ash, and high yield of toxic combustion products can be overcome by applying of a two-stage processing scheme, the gasification of a solid fuel at the first stage and further using of the fuel gas and liquid pyrolysis products at the second stage. Investigations in this direction are actively performed all over the world [1–3].

At the filtration combustion (FC), when a solid fuel and an oxidant gas move in opposite directions relative to the combustion front, the fuel is preheated and subjected to pyrolysis. As a result, droplets of liquid products (tars) and gaseous products form a combustible mist. Depending on a set of control parameters, the thermal structure of the combustion wave and the combustion temperature can vary widely. The yield of pyrolysis products in different combustion regimes can also vary considerably. The measurement of the yield and the composition of products of the thermal destruction of solid fuels become important points for studies. The liquid hydrocarbons formed at the pyrolysis of solid fuels can become a promising feedstock for new processes of conversion into valuable chemical products.

The method of the FC in the regime with superadiabatic heating is proposed for the gasification of solid fuels [4]. The gasification of solid fuels is accompanied by the formation of syngas components and pyrolysis tars. The syngas might be used for making of liquid hydrocarbons using the Fischer-Tropsch synthesis. But the liquid products should be processed additionally via hydrogenation or hydrocracking to be converted into jet kerosene or diesel fuels.

The method for gasification, the FC with superadiabatic heating, allows performing in a single reactor both the gasification and the pyrolysis processes [4]. The method is distinguished by a high chemical efficiency (up to 90%), a possibility to use fuels with high ash content (up to 95%) or high humidity (up to 70%). It does not require a complicated and costly gas cleansing system (the formation of toxic combustion products is reduced by 10–100 times compared with direct combustion). An important advantage of this process is that relatively cheap substandard fuels and some industrial wastes can be used. Conversion of these fuels and wastes to synthetic liquid and gaseous fuels, which are more convenient for further use, can be economically and ecologically favorable. In this case, valuable oil and natural gas could be replaced by local and much cheaper fuels.

The object of this study was to determine yields of gaseous and liquid products of the gasification by FC for several typical solid fuels. Effects of the fuel particles size, the ash content, the humidity, and other parameters on yields of products of the gasification were studied.

2. The Choice of Objects

Before starting of the research it is expedient to consider some advantages and disadvantages of the solid fuels and industrial wastes, in order to highlight ones most suitable for the production of liquid hydrocarbons by the FC in the regime with superadiabatic heating.

2.1. Oil Shale. Technologies and processing methods for production of liquid hydrocarbons (liquid fuels or other valuable products) from shale have been developing for a long time and achieved some substantial progress. The disadvantages of oil shale are their usually high ash content (up to 70%), high content of sulfur, and other impurities. The yield of oil shale is (depending on ash content) 6 to 25% wt. [5].

2.2. Bituminous and Brown Coals. Liquid products obtained by processing such fuels have long been valuable raw material for the chemical industry as well as for the manufacturing of synthetic motor fuels. For all types of coals common drawbacks are relatively high content of harmful admixtures and low yield of liquid products formed during the processing. For both brown and substandard coals the main drawbacks are relatively high humidity and ash content. The yield of tars from bituminous coal is within 2–10%; from brown coal it is 5–15% wt. [5].

2.3. Wood and Wood Wastes. Their advantages are the reproducibility, the availability nearly in all regions, and absence of environmentally harmful admixtures. Their drawbacks are low calorific value and, as a rule, high humidity. During thermal decomposition the yield of liquid products is up to 70% wt.; however, formed tars contain up to 30% of moisture and up to 7% of organic acids. This can lead to the corrosion of certain parts and components of the equipment [1].

2.4. Peat. Advantages of peat include its availability, relatively low contents of ash, and environmentally harmful admixtures. It also could be considered as a renewable resource. The calorific values of peat and liquid hydrocarbons formed from it are slightly higher than that of wood and wood tars. Peat tars can be a valuable chemical feedstock; however, their yield is significantly lower than the yield of wood tars (8–12% wt.). The main drawback is the high natural humidity of peat, and it may be also noted that formed products (organic acids, ammonia) are very aggressive. On top of that the ash of many types of peat is low melting [6].

2.5. Rubber Wastes. Rubber is widely used in everyday life and industry. Currently, various technologies for processing of rubber wastes are developing very fast. Thermal processing of rubber gives a high yield (~50%) of sufficient quality liquid
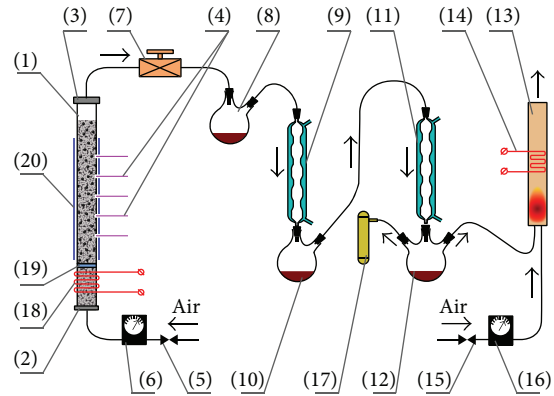

Figure 1: Schematic of the experimental setup. (1) Reactor; (2), (3) upper and bottom flanges of the reactor; (4) thermocouples; (5), (15) valve; (6), (16) flowmeter; (7) tar separator; (8), (10), and (12) flasks for liquid products; (9), (11) water coolers; (13) afterburner; (14), (18) electric heaters; (17) sample of gaseous products; (19) igniter composition; (20) radiation screen.

hydrocarbons. The main drawback (which is typical for all products containing sulfur) of produced liquid hydrocarbons is the presence of toxic and foul-smelling volatile compounds which cause environmental problems during thermal processing [7].

Each of these materials is available in sufficient quantity to give a feedstock for a small-scale power generation or chemical industry. The yield of produced liquid hydrocarbons at the thermal processing of these materials is amply high, and for each type of these liquid hydrocarbons, there are methods of treatment with recovering valuable chemical components and production of lubricating oils or motor fuels. A set of new improved methods are being developed currently [8–12].

3. Experimental

Figure 1 schematically presents the experimental setup. The experiments were carried out in the quartz reactor (1) of 46 mm diameter and 800 mm length and equipped by six thermocouples (4). The outer surface of the reactor was provided with a radiation screen to reduce heat losses (20). The tar collection system was attached to the top of the reactor. The description of the tar collection system is given in Figure 1.

Before the experiment crumbs of crashed chamotte bricks were placed in the lower part of the reactor and topped with an igniter composition—1.5 g of sawdust and 2 g of activated coal (19). The studied mixture of the combustible material with some solid inert material was placed atop the igniter composition. The process was initiated by the electric heater (18) which was wound around the outer surface in the lower part of the reactor. After heating the lower part of the reactor (with chamotte crumbs) to the temperature of 400–500°C, the air supply to the lower end of the reactor was started. That resulted in the inflammation of the igniter composition and further FC of the mixture under investigation. Thus, the FC wave formed and propagated upwards.

The upper end of the reactor was connected with a flange (3) and with a tube which directs the formed aerosol (a mixture of gas, steam, fine droplets of tars, and other organic substances) to a tars collection system. Three schemes of the tars collection system, which separate liquid products of gasification, were tested. The first one was a simplest two-stage cooling of the product gas using water coolers. Significant drawbacks of that scheme were a relatively low total amount of collected liquid products (less than 70% of the original mass) and that it collects mainly water (50–90%). The worst performance of tars collection was observed in the range of "reaction leading" combustion waves [4], at the gasification of high ash fuels, when the temperature of the product gas is low and the tar content in the product gas is lower than that at "reaction trailing" combustion waves.

The second scheme assumed a one-step cooling with the intensive contact of the product gas with a cold surface. In this case, the fraction of collected liquid products was lower than for the first scheme (up to 50%); however, the part of water in the collected liquid products was significantly less. This can compensate the drawback of the low yield of liquid products, because the water should have been removed before further treatment.

The third scheme of collecting of liquid products has advantages of both the above-mentioned ones and was found to be the most successful. The scheme is presented in Figure 1. It comprises a mechanical tar separator (7), whose operation is based on the throttling of the aerosol through a narrow nozzle, where the thickening of liquid products from the aerosol takes place. Gasification products from the outlet of the reactor passed consecutively through the tar separator (7) and then through two water coolers (9) and (11) in order to assess the effectiveness of the separator. The tar was mainly collected in a flask (8) after the tar separator. There was mostly water with a small content of organics in flasks (10) and (12). The third scheme allowed entrapping about 90% of liquid products.

The gas at the outlet of the liquid products collection system was burnt by an afterburner (13). Combustible gas was supplied to the afterburner through the bottom flange. The air required for gas combustion was fed into the burner through a quartz tube. The air-gas mixture was ignited using electric heater (14).

After the experiment, the solid combustion products were unloaded from the reactor. It comprised crumbs of the solid inert material (practically unchanged) and a small amount of ash. Collected liquid products were weighed, then dried in a desiccator, and reweighed. After that it was analyzed by burning in oxygen.

During each experiment, temperature profiles were being recorded and gaseous products were sampled. Temperature measurements were performed using Pt/Pt-Rh thermocouples, which were located on the inner surface of the reactor wall and in the center of reactor. The gaseous products composition was analyzed using GC-CRYSTAL 5000 chromatograph.

In our experiments FC of mixtures of the solid fuel with crumbs of the inert material (SHL-1.3 chamotte) was studied. The fraction of the fuel in the mixture varied from 10 to 100%.

The particle size of the inert material was equal to that of the fuel. The particle size of initial mixtures was 5–7 mm unless otherwise specified. The moisture content of the fuel was not more than 10% unless otherwise specified. The porosity of initial packed bed was 0.4–0.45. The studied fuels were oil shale (Leningrad deposit), brown coal (Tula region), wood, cellulose (cotton cloth), peat (Ukraine, Vinnitsa region, and Belarus, Minsk region), and rubber. Air was used as an oxidant in all experiments. The specific air flow (the volume air flow rate divided by the cross-sectional area of the reactor) was constant—680 m^3 /(h∗m^2).

4. Results

The heating rate and the temperature in the pyrolysis zone are the main factors which influence thermal decomposition of fuels. It is known that, regardless of other conditions, an increase in the temperature of pyrolysis always enhances the degradation of fuel [5]. At the same final temperature, the more rapid heating, the higher tar yield and the lower yield of char residue.

For the FC of solid fuels the yield of products significantly depends on the content of combustibles in the mixture. This is due to the fact that the regime of heating changes in the forefront of the combustion wave where fuel decomposes. This can be shown most clearly on the example of wood which we most often used in our experiments (including during the test of tar collecting system). Figure 2 shows the diagram of the adiabatic combustion temperature (curve (1)) and the temperature in the center (curve (2)) and on the wall (curve (3)) of the reactor and the heating rate of wood particles (curve (4)) versus wood content in the mixture. The temperatures at the reactor wall were significantly lower than those in the center of the reactor due to lateral heat loss. We estimate lateral heat loss in the reactor amounts to about 30% of the total heat release. First the combustion temperature rises with the increase of the wood content until it reaches the maximum value at 50–70% of the wood in the mixture. In this range effective conditions of heat recovery are implemented and the combustion temperature exceeds the adiabatic temperature. Further increase in the wood content causes slight decrease in the combustion temperature.

The heating rate of fuel was calculated as the ratio of the difference between the maximum temperature and 150°C versus time of this temperature increasing. As shown earlier in [13], the thermocouple indicates the temperature of the solid phase, so we use these values to calculate the heating rate of fuel. The heating rate of the fuel (Figure 2, curve (4)) invariably decreases from approximately 700 to 100 degrees per minute as the wood content increases from 10 to 100%.

When the content of the wood is low (under 20%), it heats fast and the ultimate temperature is relatively low, so the bulk of the wood changes into a slightly charred wood coke with a relatively high content of oxygen and hydrogen. After getting into the combustion zone, it mainly converts to CO_2 and water. The amount of liquid products formed in this case is relatively low, 46–52% (see Figure 3). As the wood content in the mixture grows, the temperature in the combustion front also grows and the heating rate subsides,
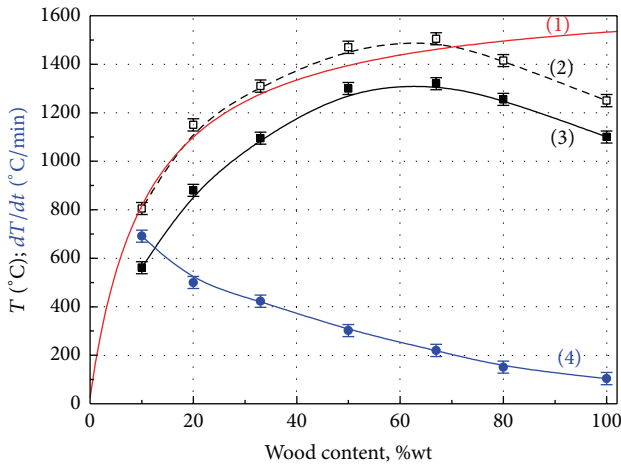
FIGURE 2: The adiabatic combustion temperature (1) and the temperature in the center (2) and on the wall (3) of the reactor and the heating rate of wood particles (4) versus the wood content.
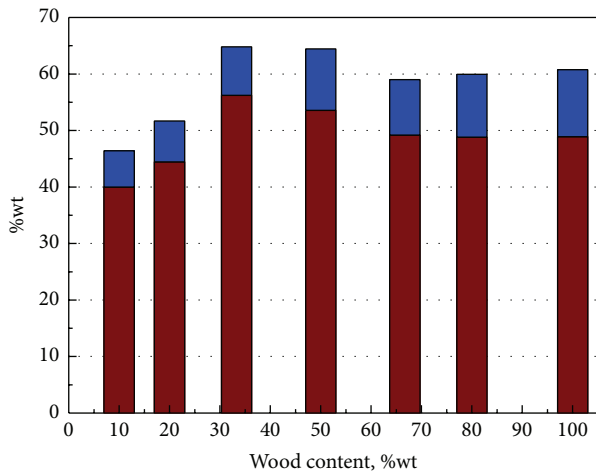

FIGURE 3: The yield of liquid products at the FC of wood. The brown column, in flask (8), the blue column, in flasks (10) and (12).

so the pyrolysis of the wood proceeds deeper and the yield of the coke decreases. The contents of hydrogen and oxygen in the coke also decrease; thereby the amount of water formed during the coke combustion decreases too. The yield of liquid products passes through the maximum of ~65% of the initial wood mass at the wood content about 30%. With the further increase in the wood content the charcoal formed becomes almost pure carbon and its yield does not change. The yield of liquid products in this range of wood content also remains almost constant and equals 60%.

The liquid products contain heavy tars, most soluble tars, an amount of other organic substances, and water. The main part of liquid products condensed when it passes through a narrow aperture with a high speed (in the turbulent regime) and then collected in a two-necked flask (see Figure 1). Next, the gas entered into the water coolers, where the condensation of remaining soluble tars occurred as well as other organic compounds (mainly acetic acid) and water.

Liquid products condensed in the water coolers poured into flasks as an acidic water emulsion. The effluent gas flowing out from the flask was almost free of tars, but it certainly contained some quantity of water vapor. The brown columns in Figure 3 show the yield of liquid products collected in flask (8), and the blue columns that in flasks (10) and (12) at the gasification of wood.

The mechanism of thermal decomposition of oil shale, peat, coal, and rubber is similar in many respects to that of wood, in spite of the fact that the physicochemical properties for all these materials are obviously different, as well as the quantity and the quality of products formed at their pyrolysis. This is due to the fact that all these materials are high-molecular compounds. The combustion temperature and the heating rate of the fuel can be changed within wide range by varying of the fuel content as in the case of wood.

Table 1 presents the yield of pyrolysis tars from various fuels (percentage of the initial weight of the fuel) at their thermal processing according to literature data. The highest yield of liquid products occurs in pyrolysis, since this process is carried out in the absence of oxygen, and, as a rule, at high heating rates. When solid fuels undergo direct gasification, the yield of the liquid products is low and in this case combustible gases are the main products. When solid fuels are gasified in a filtration mode, the yield of liquid products is comparable to that obtained in pyrolysis. It is due to (1) a self-localization of pyrolysis and oxidation zones in the reactor in the filtration mode and (2) a possibility of varying the heating rate of the fuel in a wide range. Usually external heat carriers and/or heat exchangers are used for pyrolysis, which leads to a significant complication of the construction and reduces the heat exchange efficiency. In the filtration mode both the pyrolysis and the gasification of the fuel take place in the reactor together, and the heat transfer between products and reactants occurs as efficient as possible without external heat exchangers.

The effect of the heating rate on the yield of products of the thermal decomposition is most remarkable in the processing of rubber, plastics, and "young" solid fuels (cellulose, wood). A general trend for all these fuels is the more the carbonization of a fuel, the less the influence of heating rate on the quantity of formed products. The yield of liquid products also reduces. This is due to the fact that organic macromolecules of more carbonized solid fuels have higher thermal stability. Table 1 shows the data on tars yield from wood, shale, peat, and lignite; the tars yield decreases with the growth of carbonization of the fuel.

The particles size of processed fuels has a significant effect on the yield of the pyrolysis products. As a rule, the larger the particles size of the fuel, the less the tars yield [5, 6, 14]. Usually fuels have low thermal conductivity, so the outer surface of particles heats up faster than the inner part. Tars formed inside particles pass towards the outer surface through more heated zones and partially decompose with the formation of the "secondary" solid, liquid, and gaseous products. In addition, products of the pyrolysis of larger fuel pieces stay longer at higher temperatures than pyrolysis products of smaller ones.

TABLE 1: The yield of tars formed from different materials at the thermal processing, %.

Fuel	Gasification[*]	Pyrolysis[*]	FC
Shale commercial (Slantsy)	17–22	17–22	20
Shale substandard	Not available	Not available	2-3
Wood	12–18	70	65
Cellulose (cotton cloth)	Not available	12–100	30–36
Peat	10	10–12	12–14
Brown coal (Tula region)	7–9	5–14	8–10
Rubber	Not available	20–45	40–50

[*]Literature data [2, 5, 6].

TABLE 2: The composition of gaseous products in air gasification of various fuels.

Fuel	Gaseous products composition, % vol.						Heating value, MJ/m^3	Gas yield, m^3/kg
	CO_2	Ar	N_2	CO	CH_4	H_2		
Wood	7.9	0.7	57.6	25.7	2.2	5.9	5.0	1.1
Cellulose (cotton cloth)	16.4	0.7	59.1	16.0	2.2	5.7	3.5	1.4
Peat (Vinnitsa)	11.5	0.7	60.5	21.1	0.3	5.9	3.4	1.3
Peat (Belarus)	8.0	0.7	58.3	23.3	0.9	8.8	4.2	1.6
Brown coal (Tula)	7.6	0.8	64.1	21.9	1.1	4.7	3.6	2.3
Shale substandard	14.3	0.9	76.9	1,9	0.2	0.3	0.4	0.6
Shale commercial (Slantsy)	24.4	0.7	63.1	7.6	1.2	3.0	2.0	0.9
Rubber	10.2	0.9	71.8	12.9	0.5	1.7	2.0	2.3

At gasification, these trends should be the same, but there are much less studies in this area than for pyrolysis. It should be taken into account that at the FC the widths of zones of combustion and pyrolysis as well as temperature conditions therein vary significantly depending on the particles size. Figure 4 presents the results of our experiments on the FC of wood. One can see a general trend: with decreasing of the particles size, the yield of liquid products increases. Thus, for wood content of 33% with the decrease in the average particles size from 13 to 4 mm the yield of tars increased by a factor of 1.5, and for wood content of 100% the yield of tars increased by the factor of 1.2 for the same decreasing in the average particles size. Figure 4 shows that at the gasification of low ash fuels the effect of particles size on the tars yield was somewhat weaker than that of high ash fuels, the elemental composition of tars remaining almost the same.

The gas formed in FC was low caloric with the combustion heat within 2–5 MJ/m^3; its combustion in a burner of the simple design did not cause any difficulty (see Table 2). The combustible gas contains a large amount of the carbon monoxide and hydrogen which are components of syngas. The output of gaseous products varies from 0.6 m^3 in the case of substandard shale to 2.3 in the case of coal and rubber per 1 kg of initial fuel.

Table 3 shows the elemental composition of some original fuels and tars formed from them. We analyzed air-dry fuels; the tars composition obtained from wood and cellulose before their drying is given here. The tars composition from shale, peat, and brown coal was determined after tars dewatering. Liquid products of the rubber decomposition are almost free of water, and therefore they were analyzed as they
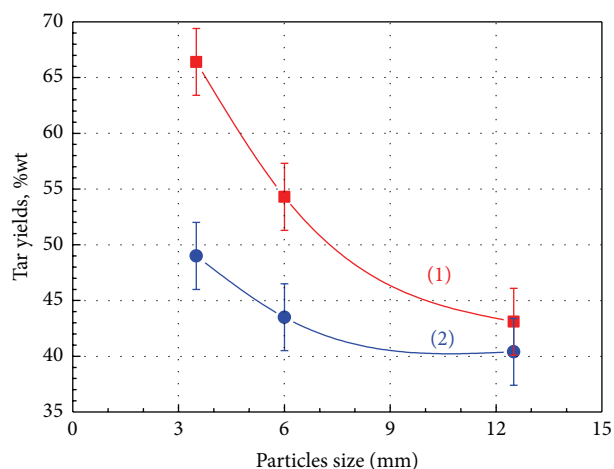
FIGURE 4: The yield of liquid products at the FC of wood. At the wood content in the mixture 33% (1), and at the wood content of 100% (2).

were obtained. The heating value of tars is high enough and varies from 18.4 MJ/kg in the case of wet tars from cellulose to 36.2 in the case of tars from shale.

The elemental composition of tars obtained from the samples studied after their drying or after draining of water almost does not depend on the temperature of FC. The general trend is clearly seen; the carbon and hydrogen content in tars increase and those of oxygen decrease compared to the original fuels. The rubber does not fit this trend and the elemental composition of its tars is nearly the same as

TABLE 3: The elemental compositions of the original fuels and tars formed from them, % mass.

Fuel	Fuel composition, %						Tar composition, %						Q, MJ/kg	Note*
	C	H	O	N	S	Ash	C	H	O	N	S	Ash		
Wood	49.8	6.4	43.2	0.1	—	0.5	52.6	6.5	40.9	—	—	—	20.0	W
							67.5	7.8	24.7	—	—	—	28.1	D
Cellulose (cotton cloth)	43.4	6.0	48.6	—	—	2.0	49.6	6.1	43.0	—	—	1.3	18.4	W
							63.7	7.4	26.9	—	—	2.0	26.2	D
Peat (Vinnitsa)	31.5	4.9	23.7	2.8	2.1	35.0	64.7	8.0	12.5	6.1	2.1	6.6	28.9	S
Peat (Belarus)	41.7	5.1	30.6	4.4	1.1	17.1	71.1	9.1	13.2	5.1	1.1	0.4	32.0	S
Brown coal (Tula)	39.2	3.2	20.2	1.5	2.3	33.6	75.4	6.8	10.6	1.2	2.9	3.1	31.6	S
Shale substandard	7.2	0.9	6.7	—	0.2	85.0	79.2	8.9	9.7	—	0.4	1.9	34.9	S
Shale commercial (Slantsy)	46.6	5.8	7.2			40.4	80.6	9.8	9.6	—	—	—	36.2	S
Rubber	84.9	7.6	2.1	0.9	1.8	2.7	84.0	6.0	4.5	1.5	2.0	2.0	34.3	W

*W: wet tars, as collected; D: tars after drying; S: tars after draining stratified water (settling for 24 hours).

that of the original rubber. It should also be noted that after tars drying the carbon and hydrogen content in tars became higher and those of oxygen decreased. This indicates that the tars originally have high water content.

Summarizing the results of our study, we can note that obtained liquid products have smaller volume and lower ash content, and their calorific values are often considerably higher than those of the original fuel. However, the content of combustibles of the liquid products is lower while the oxygen content and the moisture are higher compared with petroleum. The typical composition of petroleum products used in industry as a liquid fuel is the following: C = 83.7–87.1%; H = 10.1–12.1%; O ≤ 2%; S = 0.3–4.4%, ash ≤ 0.5%; moisture ≤ 1.5% [15]. The composition of coal and shale pyrolysis tars differ from petroleum in some lower content of hydrogen and higher content of oxygen. To make the tars obtained from solid fuels usable as motor fuels one needs to enrich them with hydrogen and remove oxygen, nitrogen, and sulfur from them. Of all the materials we studied, only liquid products obtained from rubber have properties that are comparable to those of industrial liquid fuels. For industrial use of the liquid products obtained from other materials they require an additional processing.

5. Conclusion

The yield of liquid and gaseous products formed at the FC of shale, cellulose, wood, peat, coals, and rubber has been investigated. At the FC of solid fuels the maximum temperature may be varied within wide limits (600–1400°C) and the heating rate of fuel particles may be varied (100–700°C/min) by changing of the fuel content in the initial mixture. Varying of the temperature and the heating rate of fuels allows obtaining liquid products in the amount (up to 65% by wt.), comparable to other thermal methods, in particular pyrolysis. But in case of the FC the carrying out of the process does not require additional energy supply because of using the heat of gasification of solid fuels. Our experiments have shown that the decrease in the particles size of fuels results in increasing of the yield of tars. This effect is more expressive for high ash fuels than for low ash ones.

Optimal regimes for the maximum yield of tars have been found.

In addition to liquid hydrocarbon products that can be used as a petrochemical feedstock, a low calorific combustion gas (up to 5 MJ/m^3) containing carbon monoxide and hydrogen forms at the FC of solid fuels.

Competing Interests

The authors declare that they have no competing interests.

Acknowledgments

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