

PHYSICAL AND NUMERICAL STUDY OF REVERSED BIOMASS GASIFICATION

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Abstract:

The work proposes a method of thermodynamic analysis of experimental modes on a set of design mode. This method makethe study the influence of gasification process parameters and optimise the gas generator operation possible. This approach can be used to study the operation of reactors differing in design and power, as well as to compare them with each other. The experimental results will be useful for validation of mathematical models. For the runs, complete data are given, sufficient for compiling mass and energy balances. The atypical modes of biomass gasification are attractive from a practical point of view. They are characterised by a compact reaction zone, high efficiency, low tar yields and stability of the gas composition with changes in reactor power.

Keywords: biomass gasification; reactor; synthesis gas; solid fuel.

دراسة فيزيائية ورقمية لتغوية الكتلة الحيوية المعكوسة

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الملخص:

يقترح العمل طريقة التحليل الديناميكي الحراري للأنماط التجريبية على مجموعة من أوضاع التصميم، مما يجعل من الممكن دراسة تأثير معاملات عملية التغوية وتحسين تشغيل مولد الغاز. يمكن استخدام هذا النهج لدراسة تشغيل المفاعلات المختلفة في التصميم والقوة، وكذلك لمقارنتها مع بعضها البعض.

كلمات مفتاحية: تغوية الكتلة الحيوية، مفاعل، غاز تخليقي، وقود صلب.

Introduction

Biomass is a promising renewable energy source. Its advantages compared to traditional fuels are as follows: biomass is considered as a carbon-neutral fuel, which allows reducing carbon dioxide emissions; its use can contribute to the energy security of energy importing countries, reducing their dependence on fossil and nuclear fuel supplies, increasing the energy use of biomass is an additional factor of economic support for agricultural regions [1]. Moreover, biomass is promising for the production of electricity and motor fuels in areas remote from power supply and transport [2, 3].

The peculiarity of biomass is low density of its distribution over the territory, which limits the economically efficient radius of its collection and causes interest in power installations of small unit capacity [4].

One of the promising ways of biomass processing is gasification of solid fuels. This technology is not new, but since the early 2000s, there has been a renewed interest in gasification due to the following factors: the tasks of reducing carbon dioxide emissions and improving the environmental friendliness of the energy sector as a whole; a significant increase in the cost of hydrocarbon raw materials.

Medium and small unit power reactors are used in biomass gasification. Among them, reactors with affixed fuel bed, characterised by simplicity of construction, are promising.

Despite the existing potential, gasification technologies have not yet been widely commercialised [5], and the main source of energy produced from biomass is the direct combustion process [6]. This situation is due to a number of technological problems specific to gas-generator plants [7, 8]. The study of the technological chain of biomass processing has shown that the main difficulties are associated with the stages of gasification and gas purification [8].

Insufficient stability of gasification modes leads to interruptions in the operation of gas generating stations and causes their low availability. The cost of gasification technologies increases due to the need to purify gas from tar and particulate matter, which is required for most types of gas consuming equipment.

The gas-generator plants are characterised by a relatively small efficiency factor in the conversion of chemical energy of fuel into chemical energy of gas obtained from it. This indicator often does not exceed 70%.

In order to improve the efficiency of gasification, additional research is required. The most rational and effective way of such research is the combination of physical experiments with the mathematical simulation. This work is devoted to the issues of

increasing the efficiency of bed gasification of biomass in a reactor of small unit capacity.

Purpose

To study the possibilities of achieving optimal conditions for gasification of biomass (solid fuel) from a thermodynamic point of view.

Research Methodology

Material and energy balances of the gasification process, compiled according to the laws of conservation of matter and energy, serve to assess the error of experimental determination and calculation of derived quantities, the direct measurement of which is difficult.

For continuous reactors, measurements are carried out when the conversion process reaches a steady-state (stationary) mode, when the recorded values, such as the composition of generated gas and temperature in the reactor, change insignificantly over time. Establishment of the steady-state mode can be preceded by the stages of heating and stabilisation of temperature in the as well as, the transient process of fuel gasification [9]. In the conducted series of experiments on charcoal and wood, the time of mode establishment after setting the set blast flow rate was 40-120minutes.

When studying the chemical processes, there are tasks for constructing both a general mass balance, made for the entire flow of substance entering and leaving the reactor, and balances for each chemical element. For a steady-state process, the elemental balance equation can be represented in the form:

$$\sum_i(m_i\chi_i^k) = \sum_j(m_j\chi_j^k) + Z^k(1)$$

where m_i and m_j – time average mass rate and yield of separate flows of substances, respectively, division into flows is made conditionally and depends on the form of element location; χ_i^k, χ_i^k – average values of mass fraction of element k in the corresponding flow; Z^k – absolute value of imbalance, taking a positive value when the total yield of element k is underestimated relative to its total flow rate, and vice versa. The value of Z^k does not only depend on the error of individual measurements, but also on the stability of the conversion process as a whole. A high frequency and duration of measurements are required to describe an unstable process.

According to equation (1), the elemental balances of the solid fuel gasification process require finding the average chemical composition and average flow rate of each of the material flows.

The following input flows must be considered: fuel, including moisture, organic and mineral parts; blast, which is a gas or steam gas mixture that may contain tar in gasification schemes with synthesis gas recirculation. Output flows are represented by: dry tarless gas; liquid gasification products – a mixture of water and organic compounds; coke and ash.

The data required for the material balance is not always complete due to difficulties in measuring a particular parameter. There are difficulties in determining the flow rate of gas, due to the presence of tar which leads to distorted readings and failure of flow meters. In the case of using the blast containing gaseous nitrogen, the required value of synthesis gas flow rate can be found from the nitrogen balance [9-11].

Assuming that the gaseous nitrogen of the blast is chemically inert and takes part in the gasification process only as a heat carrier, and the only nitrogen-containing product of the thermochemical

transformation of fuel is N_2 , the balance equation can be written in the form:

$$V_d Y_d^N (M^N / \nu) + m_{pt} \chi_{pt}^N = V_{cg} Y_{cg}^N (M^N / \nu) + Z^N, \quad (2)$$

where V_d and V_{cg} – blast and dry gas flow rate (m^3/h); Y_d^N and Y_{cg}^N – nitrogen fraction in blast and dry gas (% volumetric); M^N – molar mass of nitrogen (kg/mol); ν - molar volume of ideal gas ($m^3/kmol$); m_{pt} and χ_{pt} - working fuel flow rate (kg/h) and nitrogen content in it (% mass). When solving this equation, the imbalance Z^N is equated to zero.

It is acceptable to neglect the nitrogen consumption with fuel if its share relative to the total flow is small and comparable to the error in determining the nitrogen consumption with blast [12].

Thus, the fraction of nitrogen entering with fuel is 0.64 and 0.16% for air gasification of wood, the combustible part of which contains 0.6% of nitrogen by mass, with the air flow coefficient of 0.2 and 0.8, respectively. In the case, when the share of fuel nitrogen cannot be neglected, it is possible to determine the air flow rate by argon balance.

Depending on the composition of the measured values, the nitrogen balance can be used to specify the nitrogen content in the gas or blast, as well as to calculate the blast flow rate [13].

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In the operation of a gas generator with a stationary layer, the fuel flow rate is not directly controlled and is a derived quantity depending on the blast flow rate, thermal mode, fuel properties and other parameters. The measurement of the flow rate is to determine

the time required for a mass of fuel to pass through the reactor cross-section. Such a measurement may be made by the volumetric shrinkage of the fuel suspension over a certain time interval, or may be organised by means of a fuel metering device serving to continuously control the bed height. Alternatively, the fuel flow rate can be found from the carbon balance [14]:

$$m_{pt}\chi_{pt}^C = V_{cg}Y_{cg}(M^C / \nu) + m_k\chi_k^C + m_g\chi_g^C + Z^C, \quad (3)$$

m_k and m_g – consumption of coke and liquid products (kg/h), χ_k^C and χ_g^C – carbon content in coke and liquid products (% mass). The description of other values is similar to the explanations given for equation (2).

The mineral part of the fuel has been taken as an inert substance. The balance of the mineral part of the fuel can be expressed by the equation:

$$m_t A_t^r = m_k A_k^d \quad (4)$$

From equation (4), coke consumption has been determined by the expression:

$$m_k = m_t A_t^r / A_k^d. \quad (5)$$

Ash content of dry coke ash residue both in experiments on charcoal and in experiments on wood has comprised 71...74%. Coke yield has been negligibly small and made from weight of working fuel 0,12 ... 0,13% for experiments on charcoal and 0,12 ... 0,14% for experiments on wood. Small yield of coke residue and organic compounds in the composition of liquid conversion products allow

neglecting the corresponding components of the balance equation for carbon (3).

The hydrogen or oxygen balance can be used to calculate the vapour content in raw synthesis gas when the methodology of liquid sampling and analysis does not allow correct consideration of the water fraction in the products. An example of such an approach is the work on the study of biomass gasification in a circulating fluidised bed reactor [15, 16].

The nitrogen and carbon balances have been used to calculate the gas yield and fuel consumption in the experimental modes reproduced on the laboratory bench, respectively. The error in the total balance has been estimated from the total hydrogen and oxygen imbalances.

In the reactor experiments, the fuel flow rate has been determined not only by the carbon balance, but also by the fuel bed velocity. The width of the confidence interval for the relative differences in fuel consumption determined by different methods has been about 5%. Such a low error is explained by small errors in determining the gas composition, air flow rate, and fuel composition (see Table 2).

The uncertainty of the material balance is determined not only by the accuracy of individual measurements, but also by the variability of flow rates and composition over time. It is not always possible to organise direct determination of any arbitrary flow parameter, or to ensure measurement accuracy sufficient to make the balance with an imbalance value not exceeding 5%. For example, it is practically difficult to realise the measurement of the average composition of fuel entering the core of a bed reactor at a certain time interval. In particular, the elemental composition of the charcoal used for a series of experiments is unstable and varies from particle to particle. The composition data of three charcoal samples

are summarised in Table 1. The variability of the fuel composition can be explained by the unequal conditions of pyrolysis of the initial mass of wood in terms of temperature and duration.

Table 1: Elemental composition of the organic part of birch coal

Sample number	Element content,%		
	C	H	O
1	74.39	3.59	22.02
2	75.89	4.10	20.01
3	79.56	3.47	16.97

The method of nonlinear optimisation of the total balance disequilibrium on a set of linear constraints has been used to refine the fuel composition. The problem has been solved by the reduced gradient method implemented in the Premium Solver add-in of MS Excel [17].

The energy balance of the gasification process is made on the basis of the material balance. For its construction it is convenient to use the concepts and notations accepted in the normative method of thermal calculation of boiler units [18,2]. The balance equation can be written in the form:

$$Q_p+(Q_v+Q_f)=Q_g+Q_2+Q_3+Q_4+Q_5,\text{kJ/kg}$$

(6)

Where Q_g is the chemical energy of the generator gas into which the useful heat is converted; summands $Q_{2...5}$ correspond to heat losses with exhaust gases (Q_2), chemical (Q_3) and mechanical (Q_4), fuel incomplete combustion and from external cooling (Q_5).

Chemical incomplete combustion in this case is provided not by the presence of combustible components in the gas composition, but

by the tar formed. In equation (6), the available heat Q_p includes the physical and chemical energy of the working fuel, while the summands Q_v and Q_f correspond to the heat introduced into the reactor by heated air and steam, respectively.

Heat losses with material flows leaving the reactor can be estimated on the basis of data on their enthalpy. Calculation of heat losses from external cooling (Q_5) is proposed to be performed on the basis of an energy balance in which they are the closing summand [18]. The disadvantage of this method of calculation is associated with the difficulty of estimating the error of such a balance with its full convergence. The accepted methods of finding the heat losses to the environment (Q_5), based on empirical dependencies and designed for thermal calculation of medium and large capacity plants, give a high error when estimating the heat losses of a low-power laboratory bench [19].

When describing the totality of thermal processes occurring in a gas generator, it is important to distinguish between the energy balance of the reactor and the balance of the gasification process as such. The fuel layer of sufficient length can be conditionally divided into two zones - active, within which the main part of thermochemical transformations takes place, and inert. The boundary of these zones is characterised by the completion of the main gasification reactions or reduction of their rates to such an extent that the further course of the thermochemical process insignificantly affects the composition and flow rate of the gas flow filtering through the bed. When drawing up the energy balance of the gasification process, it is necessary to take into account the temperature of material flows leaving the active zone, and when drawing up the reactor balance – the temperature of flows leaving the inert zone of the bed.

Jayah et al. estimate the reaction zone height to be about 240 mm for a reversed reactor fed with 10 mm wood chips [20]. Charcoal is characterised by a smaller reaction zone height than wood. Kolodtsev's studies on air gasification of charcoal conducted on a reversed top-fired reactor indicate that conversion reactions occur within the first three particles of the bed [21]. In the experimental work by Hallett et al. on charcoal conversion in a direct-fired reactor, the height of the reaction zone was approximately five fuel particle diameters [22]. In the present study, the height of the fuel bed has been measured from the plane of the tuyere to the grate and taken as excessive. It comprised 80 mm for birch coal and 300 mm for aspen chips.

Thermodynamic simulation in this work is based on the consideration of equilibrium in the chemically reacting system. Material constraints on the yield of non-equilibrium reaction products are not used in the simulation. The equilibrium composition of the chemical system is calculated using a model with constant temperature and pressure. In terms of mathematical programming, the model has the form [23]:

It is necessary to find

$$\min G(x) = \sum G_j(x) x_j \quad (7)$$

On condition that

$$\sum a_{ij} x_j = b_i, \quad i = 1 \dots m, \quad (8)$$

$$x_j \geq 0, \quad \forall j = 1 \dots n. \quad (9)$$

Where $G(x)$ and $G_j(x)$ are the Gibbs energy of the reaction system and its j substance;

a_{ij} is the number of atoms of element i in the molecule of substance j ; b_i is the amount of substance of element i in the system, m and n are the number of substances and elements, respectively. The right part of expression (7) represents the total Gibbs energy of the reacting substances. Equation (8) sets the material balance condition and in combination with inequality (9) defines the region of physical values of variables x_j .

The mathematical model (7)-(9) can be solved in various ways. Among them, one can distinguish direct finding of the optimal value of the thermodynamic function using one or another numerical method [24], as well as approaches combining the method of uncertain Lagrange multipliers with numerical methods [25, 26], using which, the equilibrium state of the reaction system can be found by solving the following system of equations and inequalities [25]:

$$\text{When } \mu_j + \sum \lambda_i a_{ij} x_j < 0, \text{ тогда } x_j = 0, \forall j = 1 \dots n,$$

$$\text{When } \mu_j + \sum \lambda_i a_{ij} x_j = 0, \text{ тогда } x_j \geq 0, \forall j = 1 \dots n, \quad (10)$$

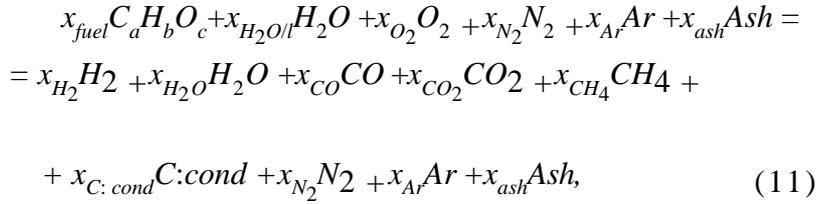
$$\sum a_{ij} x_j = b_i, i = 1 \dots m,$$

Where $\lambda_{i,j}$ – uncertain Lagrange multipliers; μ – chemical potential of the substance j (partial derivative of the Gibbs energy [27]).

The system (10) is used to establish the presence or absence of a phase in equilibrium, such as thermodynamically stable solid carbon (C: cond).

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The reaction of fuel gasification by air blowing can be written in the following form:



Where x_{fuel} and $x_{H_2O/l}$ are the amount of substance for a conditional molecule of the combustible part of fuel and water entering the process with fuel moisture, respectively. In chemical equation (11), gaseous nitrogen, argon, and ash (Ash) are taken as inert substances that do not participate in chemical reactions but require heat for their heating. Ash is modelled as a mixture of pure condensed substances. The content of ash components has been set according to [150] and made up, % of mass: $SiO_2 - 23.0; CaO - 44.5; K_2O - 7.7; Al_2O_3 - 5.3; MgO - 6.3; Fe_2O_3 - 3.6; K_2SO_4 - 6.3; Na_2O - 3.0$ and $TiO_2 - 0.3$.

$$C: x_{C: cond} + x_{CO} + x_{CO_2} + x_{CH_4} = b_C, \quad (12)$$

$$H: 2 x_{H_2} + 2 x_{H_2O} + 4 x_{CH_4} = b_H, \quad (13)$$

$$O: 2 x_{CO} + 2 x_{CO_2} + x_{H_2O} = b_O, \quad (14)$$

$$x_{C: cond} + x_{CO} + x_{CO_2} + x_{CH_4} + x_{H_2} + x_{H_2O} = x_{C: cond} + x_{gas} = \sum x_j, \quad (15)$$

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$$\Delta G^0_{CO} / RT + \ln(x_{CO} / x_{gas}) + \ln(P / P_0) + (\lambda_C + \lambda_O) / RT = 0, \quad (16)$$

$$\Delta G^0_{CO_2} / RT + \ln(x_{CO_2} / x_{gas}) + \ln(P / P_0) + (\lambda_C + 2\lambda_O) / RT = 0, \quad (17)$$

$$\Delta G^0_{CH_4} / RT + \ln(x_{CH_4} / x_{gas}) + \ln(P / P_0) + (\lambda_C + 4\lambda_H) / RT = 0, \quad (18)$$

$$\Delta G^0_{H_2} / RT + \ln(x_{H_2} / x_{gas}) + \ln(P / P_0) + 2\lambda_H / RT = 0, \quad (19)$$

$$\Delta G^0_{H_2O} / RT + \ln(x_{H_2O} / x_{gas}) + \ln(P / P_0) + (2\lambda_H + \lambda_O) / RT = 0, \quad (20)$$

$$\Delta G^0_{C:cond} / RT + \lambda_C / RT \leq 0, \quad (21)$$

$$Q_{OC} = Q_v - Q_s = \sum h_j (T) x_j - \sum h_j (T^{in}) x_j^{in}, \quad (22)$$

To find the equilibrium composition of gasification products, we solve the system of equations (12)-(22), consisting of the equations of balance by elements (12)-(14), partial derivatives of the Lagrange function for individual substances (16)-(21) and the equation of energy balance between the reaction system and the environment

(22). The last equation allows passing from the solution of the problem of searching for equilibrium at constant temperature and pressure, to the search for equilibrium at constant enthalpy and pressure of the reaction system.

In equation (22) $h_j(T)$ is the total enthalpy of its j substance, the index in denotes the initial state of the system, Q_{oc} is the heat of exchange between the reactor and the environment, Q_s is the heat loss from external cooling of the reactor, Q_v is the heat introduced into the reactor by heated air.

The relative value of Q_{os} can be found from the energy balance of a particular experimental mode:

$$q_{oc} = (Q_{oc} / Q_p) 100 = [(Q_v - Q_s) / Q_p] 100 = q_v - q_s, \% \quad (23)$$

For a process in which the blast air is not heated, the value of Q_v can be assumed to be zero. The value of q_{oc} in this case is negative and corresponds to heat losses (q_s) to the environment. In adiabatic conditions it is equal to zero, q_{oc} takes positive values when heat input to the reactor from outside exceeds its heat losses from external cooling.

The unknown quantities in the system of equations (12)-(22) are: quantities of chemical substances x_j (x_{H_2} , x_{H_2O} , x_{CO} , x_{CO_2} , x_{CH_4} , $x_{C:cond}$), Lagrange multipliers for each chemical element λ_i (λ_C , λ_H , λ_O), total number of moles of substances in the reaction system Σx_j and temperature. The number of unknowns is equal to the number of equations in the system (12)-(22).

Thermodynamic properties of individual substances were evaluated in the present work according to the data in the work [28],

atomic weights of elements were taken according to IUPAC data [29].

The method of calculation of thermodynamic properties of fuel and the specifics of representation in the coke residue model are given below.

Data of standard thermodynamic functions of individual substances are widely used in various standard reference databases.

On the contrary, the thermodynamic properties of solid fuels, as a rule, are calculated from semi-empirical dependences, which often turn out to be inaccurate. In the present work, the assumption is made that the conventional molecule of the organic part of fuel has the conventional chemical formula $C_aH_bO_c$. Then the process of complete combustion of fuel will correspond to the chemical equation:



Based on equation (24), the enthalpy of formation of a conditional fuel molecule can be estimated by the expression:

$$\Delta_f H(298)_T = Q_s^{daf} + \Delta_f H(298)_{CO_2} + 0,5x \Delta_f H(298)_{H_2O} \quad (25)$$

Where $\Delta_f H(298)$ is the standard enthalpy of formation of the corresponding substance, kJ/mol; Q_s^{daf} – higher heat value of fuel, kJ/mol. The higher heat value of solid fuels calculated using the equation proposed by A. Channiwala [30]:

$$Q_s^d = 0.3491C + 1.1783H - 0.1034O - 0.0151N - 0.0211A, \\ MJ/kg, \quad (26)$$

Where C, H, S, O, N, A – content of chemical elements and ash in fuel, %. Conversion of heat value from dry to dry ash-free mass is carried out according to the equation:

$$Q_s^{daf} = Q_s^d [100/(100 - A^d)] \quad (27)$$

The values included in the right part of equation (26) are determined by methods of elemental and technical analysis. This equation is universal for fuels of different composition and takes into account the thermal effect of ash formation from mineral components of fuel. The average absolute error of determination of the higher heat value by this method is relatively small and amounts to 1.45 %.

The standard entropy of formation of a conditional molecule of the organic part of fuel is calculated according to the method adopted for fossil coals [31]

$$\Delta_f S(298)_T = a_1 + a_2 \exp[-a_3(H/(C + N)) + a_4(O/(C + N)) + a_5(N/(C + N)) + a_6(S/(C + N))], \text{ J/mol/K} \quad (28)$$

where a_x – empirical coefficients ($a_1 = 37.1653$, $a_2 = -31.4767$, $a_3 = 0.564682$; $a_4 = 20.1145$; $a_5 = 54.3111$; $a_6 = 44.6712$) and C, H, O, N – coefficients included in the chemical formula of the organic mass of fuel. Entropy of the organic part of fuel is calculated by the equation:

$$S(T)_T = \Delta_f S(298)_T + \int_{298}^T (Cp(T) / T) dT. \quad (29)$$

Isobaric heat capacity of fuel was estimated as a function of temperature and volatile matter yield by expressions [32]:

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$$T < 400 \text{ } ^\circ\text{C}: C_p = 0.83(1 + 0.08V^{daf})[1 + 0.2T / 100 - 0.0033(T / 100)^3], \quad (30)$$

$$T > 400 \text{ } ^\circ\text{C}: C_p = C_p(400 \text{ } ^\circ\text{C}) + 0.00473(T - 400)^{0.726}. \quad (31)$$

The heat capacity has the dimension kJ/kg/K, the volatile yield is given as mass fraction. Equations (30) and (31) are used for different temperature ranges. This is due to the temperature stability of fuel. Expression (30) reflects the properties of solid fuels, while expression (31) reflects the properties of pyrolysis products.

Results and discussion

The experiments have been carried out on a laboratory bench (Fig. 1). The bench includes an inverted reactor without a neck with an inner diameter of 80 mm.

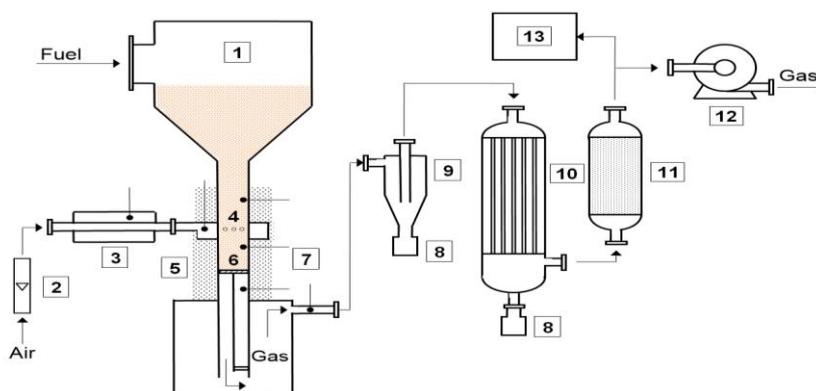


Figure 1 – Schematic diagram of the experimental stand:

1 – fuel hopper; 2 – rotameter; 3 – blast heater; 4 – tuyeres; 5 – heat insulation; 6 – grate; 7 – thermocouples; 8 – condensate receivers; 9 – cyclone; 10 – heat exchanger; 11 – fine filter; 12 – exhauster; 13 – chromatograph.

The maximum thermal power of the reactor in the conducted experiments has comprised 8.5 kW.

The experimental works have had the following number of features:

1. Fuel consumption in the studied process should be a derived parameter and determined by the proper needs of the gasification process. For this purpose, a bed process without forced removal of the bed from the reactor has been chosen.

2. In order to avoid errors associated with underestimation of tar yield in the thermodynamic models, a reversed gasification process has been chosen to achieve deep tar decomposition. For stepwise validation of the model, charcoal gasification modes characterised by lower volatile methane yields have been used. Methane yields, like tar, are also underestimated in thermodynamic models.

3. It is necessary to study the process parameters that are not taken into account directly in the model. In the experiments one such parameter has been changed, namely the flow rate of the gasifying agent.

4. The heat exchanged by the reactor with the environment should change in a wide range of values. In the experiments, the value of reactor heat loss from external cooling depended on the thickness of thermal insulation, and the heat input from outside has been achieved by heating the gasifying agent.

Fuel is supplied to the reactor from the hopper (1). Air is supplied to the reactor through an electric heater (3) and six tuyeres (4) placed evenly around the perimeter of the reactor. The reactor has a thermal insulation layer (5). The fuel bed rests on a grate (6), which can be set at a particular height within the reactor. The grate allows setting

the bed height, which varies from the minimum (grate opposite the tuyeres) to 455 mm.

The temperature measurements are carried out using K type thermocouples (7), whose jaws are located near the inner surface of the reactor. This arrangement of thermocouples does not prevent fuel shrinkage. Solid particles are separated from the generator gas in the cyclone (9). The cyclone also allows the collection of liquid products. The gas from the cyclone is cooled in a heat exchanger (10) and passes through a fine filter (11). The gas is pumped through the unit by a fan (12). It creates a discharge inside the unit in the order of 2...6 kPa, thus excluding leakage of generator gas into the laboratory premises. Air flow rate is controlled by the fan power and measured by rotameter (2).

The gas composition has been determined by gas chromatography. Chromatograph SRI8610C, equipped with silica gel and 13X zeolite columns connected in series, allows the determination of all gas components, including C₂-hydrocarbons. With helium as carrier gas, the analysis time is about 12.5 minutes. The sum of absolute errors of gas component concentrations does not exceed 1%.

The yield of liquid gasification products has been estimated by weight of samples taken from the cyclone and heat exchanger. The tar content in the samples has been determined by the weight of residue obtained after drying a part of the averaged sample (10 ml) in a desiccator at 70 °C for 40...50 minutes. A similar approach was used earlier in the work of Bhattacharya and Dutta [33].

Aspen chips with particle size 9...11 mm and chopped birch coal with particle size 8...12 mm have been used in the experiments. Before loading the fuel into the hopper, it has been thoroughly mixed and a sample has been taken for analysis. The data of elemental and technical analyses of the fuels are presented in Table

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1. The table also summarises the properties of aspen coal obtained in one of the experimental runs. Unlike wood, the charcoal has not been stored in a hermetically sealed container and its moisture content varied from run to run.

The parameters measured during the experiment and the accuracy of their determination are summarised in Table 2.

Table 2: Measurements in the experiment

Measured value	Measurement method	Error
Air consumption	Float-operated rotameter	$\pm 2.5\%$
Air humidity	Moisture meter VIT-2	$< 5\%$
Air and generator gas temperature	Cr/Al thermocouple, ADC amplifiers MAX31856	$\pm 10^\circ \text{C}$
Chemical composition of gas	Gas chromatographs SRI8610 CandInficon Fusion	$< 1\%$
Elementary fuel composition	GOST2408.4-98	$< 0.5^*\%$
Humidity, yield volatiles, ash content	GOST27589-91,55660-2013,11022-95	$< 0.5^*\%$
Consumption of liquid products	by sample weight and time of collection	–

Note: *—method reproducibility.

In the reversed gas generators, it is recommended to use biomass with humidity not exceeding 20 % [34]. If the fuel bunker is heated with generator gas and in multizone reactors, it is possible to use fuel with humidity up to 30...45% [35]. However, this leads to a significant decrease in the heat of gas combustion and gasification efficiency. For the experiments on biomass gasification, sufficiently dry fuel containing 7% moisture has been taken (Table 3).

In the frame of a series of experiments, a mode with air blowing and excess bed height equal to 455 has been set. Figure 2

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shows data on this mode. The gas temperature has been taken from the readings of thermocouples, the hot junction of which is located in the reactor wall space. Heat losses from external cooling of the reactor, calculated by the energy balance equation, depend on the position of the point at which the temperature of the exhaust gases is measured. As the distance from the tuyere plane increases, these heat losses increase twofold due to the cooling of the fuel and gas bed filtering through it.

Table 3: Properties of fuel and coke residue

Value, % wt.	Birch charcoal			Aspen wood chips	Aspen charcoal
Launch ID	C1	C2	C3	B1-B3	B3
Operational humidity	3.6	4.4	2.6	6.8	0.9
Elemental Composition					
C	82.5			49.3	86.6
H	3.6			5.5	1.7
O	13.9			45.2	11.7
Results of Technical Analysis					
Volatile yield, V ^d	18.9			84.9	13.0
Non-volatile carbon, dry	77.6			14.7	84.7
Ash, A ^d	3.5			0.4	71.0-74.3
Flash point, °C	232			285	-

For correct construction of the energy balance of the gasification process, it is necessary to measure the temperature of the substances flows leaving the reactor core. It is possible to determine the extent of this core by determining the gas composition at a certain section along the length of the reactor. However, the study of gas formation dynamics along the bed height is a separate task, which is beyond the scope of this work.

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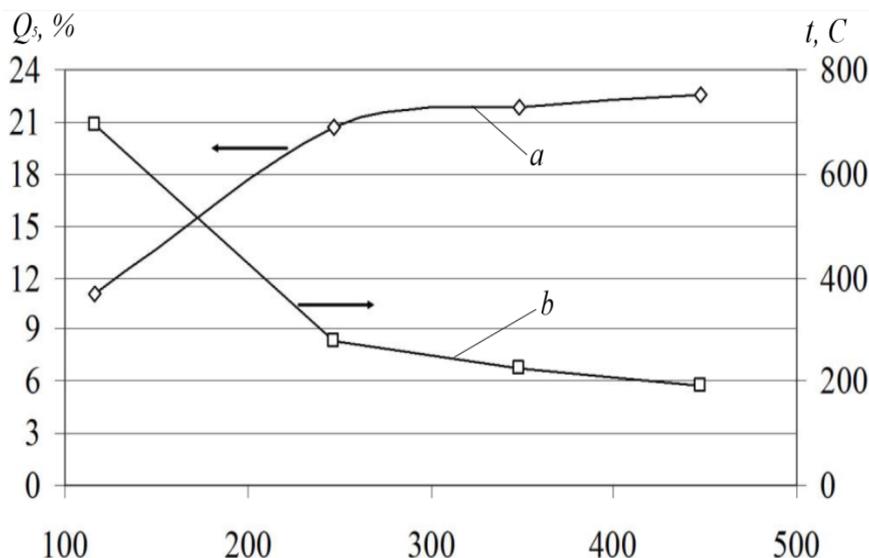


Figure 2 - Dependence of heat losses from external cooling (*a*) and gas temperature (*b*) as a function of the distance between the thermocouple and the tuyere plane

In the experiments on charcoal, the temperature of flue gases has been taken according to the readings of a thermocouple located directly in the fuel bed at a distance of 52 mm below the tuyeres. The readings of the same thermocouple have been taken in the experiments on wood, for which the dimensions of the reaction zone appeared to be comparable to those on coal.

The entropy of formation and heat capacity of fuel do not influence the results of thermodynamic calculations. Their use in the equilibrium model (12)-(22) is justified only from the position of completeness of the thermodynamic description. This is due to the absence of the initial fuel in equilibrium (Table 4).

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Table 4: Modelling of air gasification of woody biomass. Properties of the coke residue are represented by graphite (Case 1) or aspen charcoal according to Table 4 (Case 2)

Initial parameters of the model		Case 1	Case2
Air flow coefficient (α)		0.15	0.15
Heat exchanged between the reactor and the environment (Q_{oc}), kJ/mol		0	0
Biomass and air temperature, °C		25	25
Equilibrium composition of the reaction system, mol/mol			
Aspen wood ($CH_{1,49}O_{0,69}$)	0.00E+00	0.00E+00	
Charcoal ($CH_{0,23}O_{0,10}$)	-	3.93E-34	
C (graphite)	0.327	-	
CH ₄	0.043	0.226	
CO	0.356	0.508	
CO ₂	0.274	0.267	
H ₂	0.561	0.332	
H ₂ O	0.198	0.061	
N ₂ +Ar	0.580	0.580	
Temperature, °C	637	588	
S, kJ/mol/K	0.467	0.464	

Coke residue is one of the main products of gasification. Using methods (24)-(31) it is possible to replace graphite with coke in the model. Such an improvement seems to be logical, since it takes into account the residual amounts of hydrogen and oxygen contained in coke. However, the thermodynamic stability of coke is less than that

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of graphite, and its equilibrium amount turns out to be extremely small (Table 4). In order to better reproduce the experimental data, the graphite yield is often corrected upwards. Substitution of graphite for coke in the model aggravates this problem and turns out to be rational only with additional use of material constraints on its actuation.

Table 5: Equilibrium composition of raw generator gas (mole fraction)

Substance	Source	Temperature, K					
		1000	1100	1200	1300	1400	1500
H ₂	Smith et al.	0.138	0.169	0.188	0.197	0.201	0.203
	Model(12)-(22)	0.137	0.169	0.187	0.197	0.201	0.203
CO	Smith et al.	0.112	0.226	0.327	0.378	0.398	0.405
	Model(12)-(22)	0.112	0.225	0.326	0.377	0.397	0.404
H ₂ O	Smith et al.	0.121	0.068	0.032	0.014	0.006	0.003
	Model(12)-(22)	0.122	0,069	0.033	0.014	0.006	0.003
CO ₂	Smith et al.	0.143	0.090	0.040	0.015	0.005	0.002
	Model(12)-(22)	0.142	0.090	0.041	0.015	0.006	0.002
N ₂	Smith et al.	0.486	0.447	0.413	0.396	0.390	0.387
	Model(12)-(22)	0.487	0.447	0.413	0.397	0.390	0.388

Verification of a thermodynamic model. Model verification is the process of establishing the accuracy with which the numerical solution obtained by a computer programme reproduces the

“conceptual” model written in the form of mathematical expressions. Unlike validation, the verification procedure does not establish the adequacy of the model’s representation of some physical object or process. Verification establishes the relation between the mathematical formulation of the problem and its numerical implementation, while validation determines the relation between the numerical solution and the modelled reality [36].

Verification of calculations is conveniently carried out with the help of repeatedly tested examples. Smith J.M. et al. present data on modelling of steam-and-air gasification of carbon obtained using the law of acting masses [26]. It is worth noting that this type of models does not take into account the air flow coefficient (α) and allows finding the equilibrium composition of the system at the carbon boundary. In the calculation, the authors vary the process temperature at a constant pressure of 20 bar (19.74 atm.). To reproduce this process, the system of equations (12)-(22) was solved, which did not include the energy balance equation. Table 5 compares the results obtained by Smith et al. and on model (12)-(22).

The absolute discrepancy in the values of mole fractions of substances is not more than 0.001, which may be due to rounding of both intermediate and final results presented in the work [26].

Conclusions

Thermodynamic analysis of experimental modes, put on the reversed gas generator, revealed three efficiency limitations. The first limitation is related to the kinetics of interaction between gas and fuel and manifests itself when their reaction time is insufficient (insufficient height of the fuel bed). In this case, the reaction system tends to reach the carbon boundary line and the maximum

conversion of available thermal energy into chemical energy of gas. Further increase of gasification efficiency is possible when the system moves along the carbon boundary line. Such motion is determined by the heat exchanged by the system with the environment. The second efficiency limitation is common to autothermal processes, which may lack their own heat. The third efficiency limitation arises when the heat supplied to the process from outside is predominantly consumed for heating substances and reactor walls. All three limitations are, to varying degrees, common to both charcoal and wood gasification processes.

The regularities of the biomass gasification process turned out to be atypical and different from the literature data. Based on the actual data, a hypothesis about the mechanism of biomass gasification has been formulated. According to this mechanism, separate stages of the process do not take place in different zones of the fuel bed, but within separate particles. The coal bed is formed on the surface of the particle reacting with oxygen with the release of heat. This heat is absorbed by heat acceptor processes. These are pyrolysis of the inner bed of biomass, processes of tar decomposition and recovery of pyrolysis products during their filtration through the surface bed of coal. This hypothesis needs further development and verification.

The experimental results will be useful for validation of mathematical models. For the runs, complete data are given, sufficient for compiling mass and energy balances. The atypical modes of biomass gasification are attractive from a practical point of view. They are characterised by a compact reaction zone, high efficiency, low tar yields and stability of the gas composition with changes in reactor power.

The work proposes an approach that allows thermodynamic analysis of experimental modes, studying the influence of process

parameters and optimising the operation of a gas generator. This approach can be used to study the operation of reactors differing in design and power, as well as to compare them with each other.

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