Reduced-Order Modeling of Pulverized Coal Staged Gasification: Influence of Primary and Secondary Fuel Proportion

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***Abstract* — The paper is concerned with the numerical simulation of the coal gasification process in an entrained flow of high-temperature air-steam mixture. Due to the high initial temperature and the process staging, it is possible to obtain an efficient gasification process. The study aims to examine the stationary conditions of staged gasification process by using a mathematical model based on one-dimensional heat and mass transfer equations with combined submodels to describe physicochemical transformations. The simulation makes it possible to determine the boundaries of the transition from the "single-stage" to the "two-stage" gasification conditions and identify the most promising ones.**

***Index Terms* — coal, staged gasification, mathematical modeling, reduced order modeling.**

I. INTRODUCTION

Gasification of solid fuels for energy gas and chemicals production is one of the promising ways to improve the technological and environmental efficiency of fuel consumption [1]. Nowadays, gasification plants are successfully operating in a number of countries around the world, and new projects are being developed for their construction [2]. Among gasifiers for large power plants (more than 100 MWe), entrained flow reactors are widely used [3]. In the entrained flow conditions, reacting mixture is a suspension of fine solid particles or droplets in gaseous oxidizer.

Gasifier usually works as part of a complex power or/and process plant [4]. Therefore, its operating conditions and parameters are determined by the operation of the plant and the power system as a whole. In this regard, the important task is to develop numerical models enabling the efficiency evaluation for different gasifier operating conditions for the comprehensive optimization of such plants.

One of the trends in gasification technologies is to increase temperature of reaction core, which improves the equilibrium yield of the target combustible components, increasing the specific rate of chemical reactions, and ensures liquid slagging. Therefore, oxygen-enriched gasification agent is most widely used in the industrial processes. Oxygen production requires additional costs, therefore, in some processes, high temperature air is used [5, 6], or a staged supply of fuel and a recycling of unreacted fuel are used [7]. According to thermodynamic estimates, the efficiency of power plant with an air-blown gasifier can be close to the efficiency of a plant with oxygen-blown gasifier [8, 9].

In [9], the authors propose a new integrated coal gasification combined cycle power plant, in which both high-temperature heating of gasification agent and recirculation of unreacted fuel were used. The present paper is devoted to the study of stationary conditions of the gasifier in a similar scheme with different ratios between primary and secondary fuel.

II. STAGED PROCESSES OF PULVERIZED COAL GASIFICATION: APPLICATION AND MODELING

The use of staged gasifiers is usually associated with the gas quenching by the utilization of process heat in endothermic chemical reactions to produce combustible products. Reduction in the temperature of the product gas allows, in general, decreasing thermodynamic losses of the process [10]. In addition, the organization of the staged gasification process simplifies the cooling of the product gas [11].

E-Gas gasifiers fed by water-fuel suspensions or emulsions are widely used in the chemical industry to produce syngas and hydrogen. In the first stage of the

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process, the fuel droplets are burned in oxygen to produce a hot gas, and in the second stage secondary fuel is injected into this gas. Fuel evaporates and gasifies due to the heat obtained in the first stage. The Wabash River thermal power plant (260 MWe) uses the E-Gas gasifier to produce fuel gas [3, 12].

The Mitsubishi Heavy Industries (MHI, later MHPS) gasifier is implemented at the Nakoso station (250 MWe) [3, 7, 13]. The fuel here is coal dust, and the primary gasification agent is air. The first stage, as in the E-Gas process, is burning of primary fuel to produce hot gas. The second stage is also the gasification of the secondary fuel in the hot combustion products. However, due to the high nitrogen content, a significant amount of coke residue is produced. Thus, the coke-ash residue is directed back to the first stage, where it is burned along with raw fuel.

The EAGLE (Energy Application for Gas, Liquid and Electricity) gasifier was developed by J-Power. The fuel conversion process in this gasifier is based on the organization of swirled flows within the reactor, when the combustion and reduction stages occur in counter-current streams within a single reactor unit. This process is presented by the pilot plant Wakamatsu (8 MW) [3].

China is actively working on the development of new gasifiers, including the staged ones: TPRI, HNCERI, etc. [14–17].

There are many papers devoted to the development of mathematical models for the staged processes of pulverized coal gasification. In some of them, the authors make attempts at CFD-modeling, which is explained by the importance of proper flow organization in the reaction zone. Thermochemical equilibrium models are often used to evaluate the operation of a gasifier as part of complex technological schemes [8, 18, 19]. The CFD-models of the E-Gas gasifier were studied and used in [20-25]; MHI gasifier models are presented in [25-32]; EAGLE gasifier models are presented in [33, 34]. The CFD-models of staged processes in laboratory and pilot units were proposed in [35–38]. The main problems of using these modelsare computational efforts and appropriate choice of empirical coefficients corresponding to submodels of turbulence and radiation transfer in dispersed flows.

The authors of [17, 39, 40] propose simplified models of staged processes, which are sets of connected one-dimensional and zero-dimensional reactors. Such models (Reduced Order Models, or ROMs) allow variants calculations and the process behavior evaluation in a wide range of conditions for a reasonable time. The present paper also uses a simplified mathematical model of a staged gasifier, which allows a sufficiently detailed study of the influence of individual factors on the gasification process characteristics with smaller computational requirements.

III. INITIAL DATA AND MODEL DESCRIPTION

Gasification process in a staged reactor can be represented as follows (see Fig. 1 and [7, 9]):

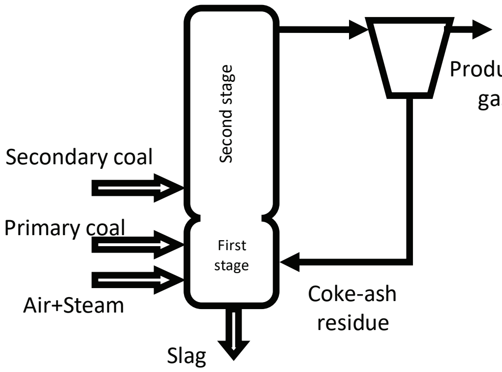

Fig. 1. Scheme of entrained-flow two-stage gasifier.

high-temperature air and steam with primary fuel are supplied to the first stage along with a coke-ash residue, which is formed in the process. Nitrogen is used as a transport media for fuel supply. Combustion occurs in the first stage, and temperature is supposed to be sufficiently high to ensure liquid slag removal. The obtained high-temperature gas enters the second stage together with the stream of secondary fuel, where its allothermic gasification occurs (for the products of the first stage are predominantly CO2 and H2O). Unreacted coke is returned to the first stage of the process. Thus, one of the important process parameters is the primary fuel proportion (i.e. primary fuel consumption divided by overall fuel consumption). This value determines the combustion stage efficiency, i.e. the first stage equivalence ratio, ?1 (equivalence ratio is defined as a ratio of actual oxygen to fuel ratio to the stoichiometric ratio). The combustion stage efficiency in turn determines gasification stage efficiency. Thus, there is a feedback that could result in specific features in the entire gasification process.

The fuel in the calculations is the Kuznetsky coal, grade D. The composition and properties of the fuel are given in Table. 1. The total length of the reaction zone is 12 m, the first stage is about 3 m; the diameter of the reactor is 3 m along the entire length. The pressure in the reactor is considered constant and equal to 3 MPa. Overall fuel consumption is constant and is equal to 70 t/h.

Preliminary thermodynamic calculations showed that the equilibrium gasification process has optimal characteristics for the yield of combustible gas under the conditions given in Table 2. For these conditions, we made calculations to find the options for implementing

Table 1. Kuznetsky coal characteristics.

Fuel characteristics	Values and units
W^r	2.9%
A^d	23.7%
C^{daf}	78.79%
H^{daf}	5.97%
N^{daf}	2.16%
S^{daf}	0.97%
O^{daf}	12.11%
V^{daf}	29.9%
d_p, μm	100

Table 2. Equilibrium parameters of kuznetsky coal air-blown gasification under 900°C

Variable	Unit	Value
Equivalence ratio, α	-	0.33
Steam ratio	mol(H_2O)/mol(C)	0.2
Cold gas efficiency	%	85.7
Producer gas composition % vol.		
CO		28.3
H_2		19.5
H_2O		2.1
CO_2		3.9
N_2		45.8

the limiting equilibrium characteristics of the gasification process.

The mathematical model used in this study was discussed in our previous papers [41–43]. This is a system of spatially one-dimensional equations of heat and mass transfer with a combined kinetic-thermodynamic submodel for solving the problem of chemical transformations. The authors previously used similar models in [39, 44–48] to estimate the characteristics of the gasification process.

The model is based on the heat balance of coal particles and the gas film adjacent to them. The following assumptions are made about the course of the gasification process [41, 42]:

1) the drying rate is limited by external mass transfer of particle with ambient gases;
2) the pyrolysis rate is proportional to the content of volatiles in the particle and depends on temperature according to the Arrhenius law;
3) the gasification rate is determined from the well-known equation of the diffusion-kinetic theory of carbon combustion.

The heat balance equation for a coal particle is written as follows:

$$c_p \frac{d\left(m_p T_p\right)}{d\tau} = \varepsilon\sigma S\left(T_w^4 - T_p^4\right) + \alpha_T S\left(T_g - T_p\right) - Q_w + Q_r.$$

Here c_p is the specific heat of the fuel, *J kg^{-1} K^{-1}*; mp is current particle mass, kg; T_p – particle temperature, K; ε is the degree of the particle blackness; σ is the Stefan-Boltzmann constant, W (m^{-2} K^{-4}); S is a particle surface area, m2; T_w is ambient temperature, K; α is convective heat transfer coefficient, W/m^{-2} K^{-1}); Q_w is heat of moisture evaporation, W; Q_r is heat of chemical reactions, W.

Drying rate is calculated by the formula:

$$j_w = K_w S\left(C_{H_2O}^{eq} - C_{H_2O}\right)$$

Here j_w is the flow of moisture, kg s^{-1}; K_w is drying rate constant, m/s; C_{H_2O} is the concentration of water vapor, kg m^{-3}.

The coefficient of convective heat and mass transfer for a particle in a stream is calculated by the formula:

$$\mathrm{Nu} = \mathrm{Sh} = 2 + 0.16\,\mathrm{Re}_p^{2/3}$$

Here Nu is the Nusselt number; Sh is the Sherwood number; Re_p is the Reynolds number for the velocity of the carrier flow and current particle size.

The pyrolysis rate is described by the first-order kinetic equation:

$$\frac{dm_V}{d\tau} = -k_V^0 \exp\left(-\frac{E_V}{RT}\right) m_V$$

Here m_v is the mass of volatiles in the particle, kg; $k^0{}_v$ – pre-exponential coefficient, s^{-1}; E_V is the activation energy of the pyrolysis stage, J/mol; R is the universal gas costant, J mol^{-1} K^{-1}. The volatiles in the model are represented by a mechanical mixture of chemical elements,. After exiting the fuel particle, volatiles achieve their molecular forms according to the conditions of chemical equilibrium.

The reaction rate of the fuel with gaseous oxidizing agents is recorded as follows:

$$\frac{dm_C}{d\tau} = -k_{eff} S C_{ox}$$

Here m_C is the mass of fuel, kg; k_{eff} is effective rate constant for heterogeneous reaction, m/s; S is a fuel surface area, m^2; C_{ox} is the oxidizer concentration, kg m^{-3}.

The effective rate constant is expressed in terms of the kinetic and mass transfer coefficients (assuming that the kinetic order of the reaction with respect to the oxidant is one) as follows [49]:

$$k_{eff} = \frac{k_C k_d}{k_C + k_d}$$

Here k_C is the kinetic rate constant for a heterogeneous reaction, m/s; k_d is the mass transfer coefficient of the particle with the flow, m/s.

The kinetic rate constant for a heterogeneous reaction depends on temperature exponentially:

$$k_C = k_C^0 \exp\left(-\frac{E_a}{RT}\right)$$

Here $k^0{}_C$ is the preexponential coefficient, m/s; E_a is activation energy, J/mol.

Overall change in particle mass is written as follows:

$$\frac{dm_p}{d\tau} = -j_w + \frac{dm_V}{d\tau} + \frac{dm_C}{d\tau}$$

Chemical kinetics of reactions in the gas phase is not considered. It is assumed that substances entering the gas phase pass into a state of equilibrium. Thus, chemical transformations are described using a thermodynamic model with macrokinetic constraints on the rate of heterogeneous transformations. This approach is applicable to high-temperature processes in which the rates of gas-phase processes are quite high compared to the rates of heterophase processes [50].

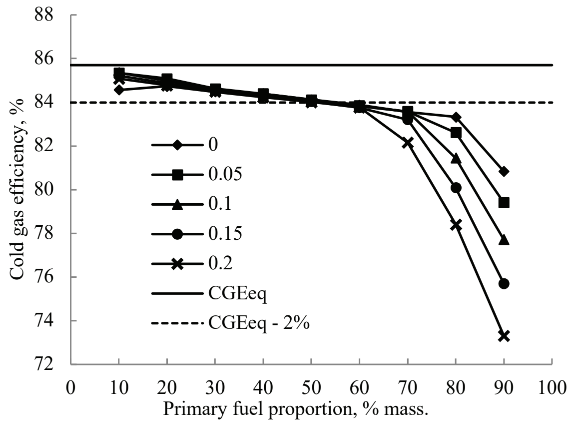

Fig. 2. Cold gas efficiency versus primary fuel and secondary steam proportions, mol(H2O)/mol(C). Dashed lines are equilibrium cold gas efficiency and 2% less values.

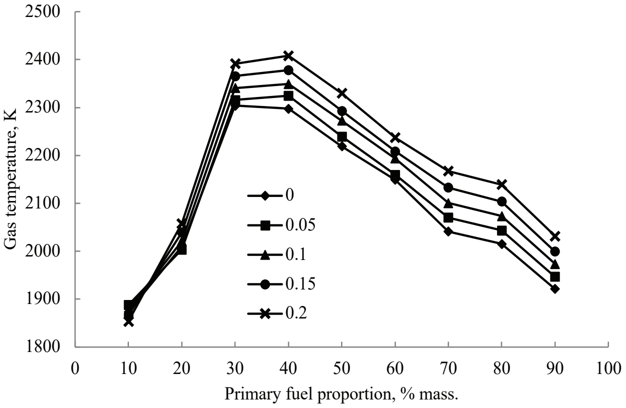

Fig. 3. The first stage output temperature versus primary fuel proportion and secondary steam proportion, mol(H2O)/mol(C).

In the model, the equations of complex heat transfer (convective and radiant) between fuel particles, reactor wall and the gas phase are solved. The wall is considered to be adiabatic. This circumstance additionally simplifies the calculations, since it allows neglecting the specific features of the heat exchange with the wall and with the cooling jacket. In a more accurate formulation, however, it is necessary to take into account the heat transfer inside the reactor wall, ash melting and thermal conditions of slag films [51, 52].

Since, as mentioned above, the transfer processes and chemical reaction are closely related, it is necessary to solve the equations of diffusion kinetics and heat transfer. In the present work, these equations are solved separately at different steps of iterative procedure. The following iterative algorithm is proposed to calculate 1) chemical transformations of fuel particles in the gas stream; 2) transfer of heat released as a result of chemical transformations. Each time chemical transformations are calculated with a refined temperature profile, then the temperature profile is adjusted to chemical transformations [53, 54].

The calculation of the staged process as a whole is carried out as follows. The algorithm is based on the repeated application of fuel particle model in a changing thermal field. This model allows us to calculate the processes in the oxidation and reduction zones separately: when combustion reactor is calculated, the output gas stream is directed to the reduction zone.

The consumption of primary fuel is fixed in each calculation. However, the total fuel consumption in the first stage varies between iterations, since the output of the coke-ash residue is not controlled. This value is set in the calculation progress. The algorithm stop condition is a small change in the total fuel consumption in the first stage: the fuel consumption in the first reactor in two successive iterations changes by less than 5%.

The feedback in the system of reactors can lead to a significant change in its behavior (as compared to the one-step process). At low degrees of secondary fuel conversion, the resulting coke-ash residue is cooled and then recycled. It leads to an increase in heat loss and to a decrease in the degree of fuel conversion in the first stage. In this

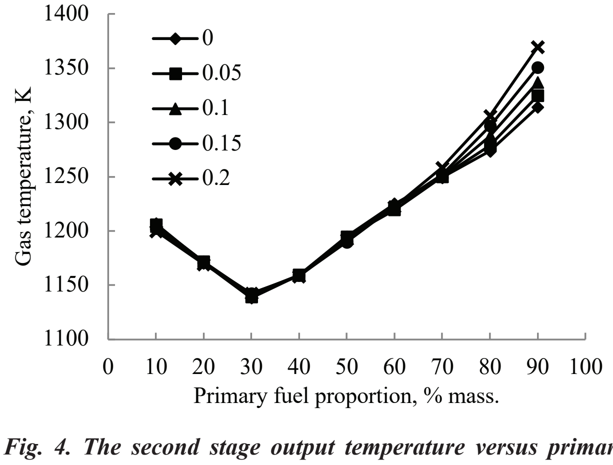

Fig. 4. The second stage output temperature versus primary fuel proportion and secondary steam proportion , mol(H2O)/mol(C).

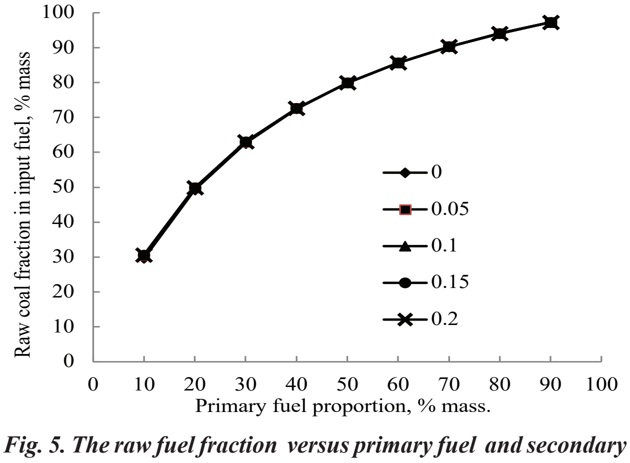

Fig. 5. The raw fuel fraction versus primary fuel and secondary steam proportions , mol(H2O)/mol(C).

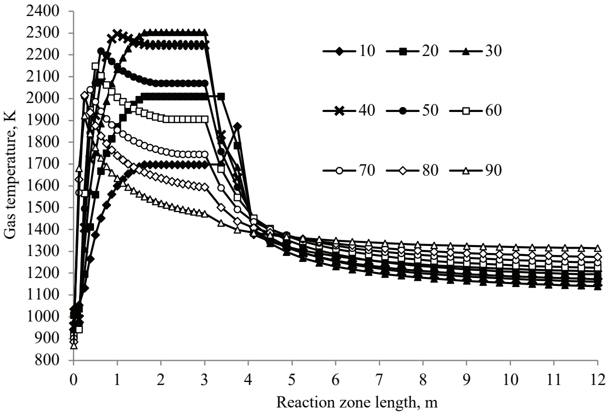

Fig. 6. Temperature distribution along reactor versus primary fuel proportion .

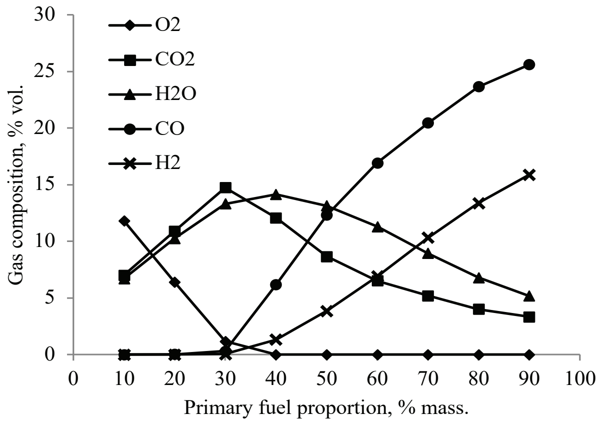

Fig. 7. The first stage output gas composition versus primary fuel proportion .

case, however, its quantity does not increase, since the calculation assumes that all the solid residue from the first stage output is carried away with the liquid slag. Actually, such operation of reactors will be ineffective. It should also be noted that, under the conditions of a stable staged gasification process, combustion temperature is to be sufficiently high to ensure liquid slag removal (otherwise, the ash will be recycled together with the coke-ash residue of the gasification stage, which is undesirable).

The model of a staged gasifier was verified in [41] for a pilot reactor with a capacity of 2 t/day. The calculations showed good agreement with the experimental data on gas composition.

IV. Results and discussion

Two fuel flows and two steam flows are considered with respect to the gasification process stages. Primary fuel and primary steam feed the first stage. Secondary fuel and secondary steam correspond to the second stage. Distribution of fuel and steam between stages is described by different proportions. The influence of these two parameters was studied numerically. Overall steam and fuel consumption is fixed. The secondary steam consumption is varied from 0 to 0.2 mol/mol(C) (the primary steam consumption is determined by the difference from 0.2 to 0 correspondingly). Primary fuel proportion varies from 10% to 90%, secondary fuel proportion changes from 90% to 10%, respectively. The results of the calculations are presented below.

Efficiency of gasification process is usually described by cold gas efficiency (CGE, ratio of producer gas heating value to solid fuel heating value). As it can be seen from Fig. 2, the cold gas efficiency is close to the equilibrium value (CGEeq) in a wide range of primary fuel ratios (up to about 50%), its further increase leads to a decrease in the cold gas efficiency due to a decrease in combustion reactor temperature (Fig. 3). At the same time, the temperature of the second stage product gas (see Fig. 4) is very sensitive to the primary fuel proportion: with its growth, the gas temperature gradually increases close to the equilibrium value (1373 K). Deviations are associated with an imbalance in reagent consumption. The low temperature of the product gas with a small primary fuel proportion is associated with heat loss due to coke-ash residue cooling. As can be seen from Fig. 5, the fraction of raw coal in a mixture with recirculating coke-ash residue could be about

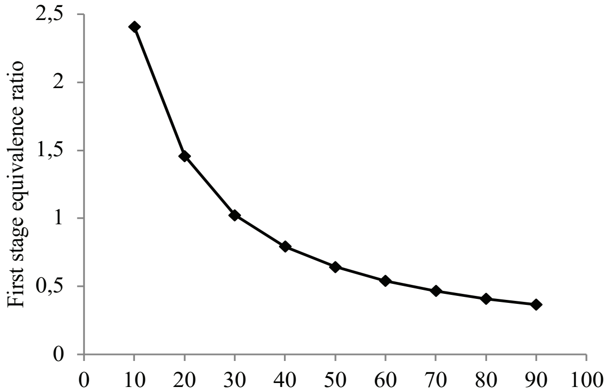

Fig. 8. The first stage equivalence ratio versus primary fuel proportion.

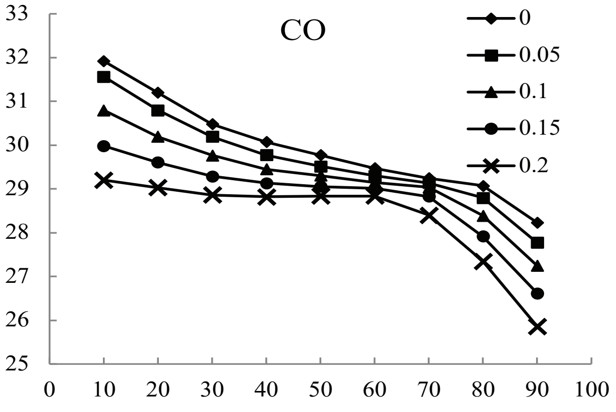

Fig. 9. The CO volume fraction in produced gas versus primary fuel and secondary steam proportions.

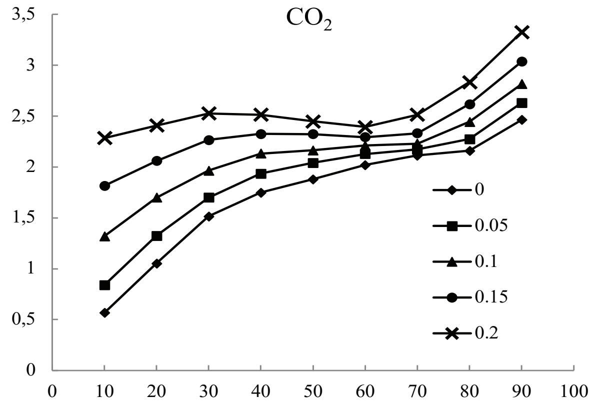

Fig. 10. The CO_2 volume fraction in produced gas versus primary fuel and secondary steam proportions.

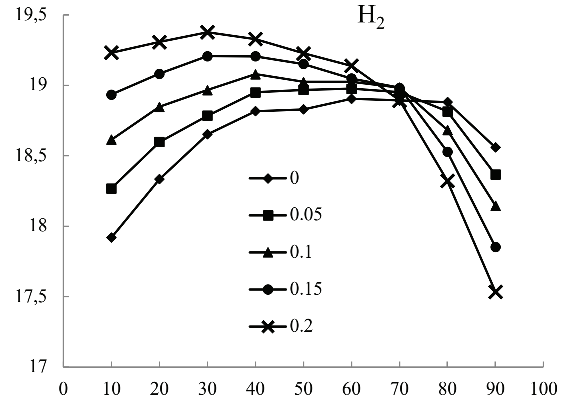

Fig. 11. of The H_2 volume fraction in produced gas versus primary fuel and secondary steam proportions.

30%. Since the model uses the assumption that the coke-ash residue is cooled during transport, and the slag is removed at the temperature of the combustion reactor, considerable heat loss occurs. Therefore, to maintain the desired level of the product gas temperature , it is necessary to deviate from the equilibrium estimate of the parameters of the optimal conditions (for example, towards an increase in the oxygen-fuel ratio).

An interesting effect is the extremum value of gas temperature: the maximum temperature of the first stage and the minimum output temperature is achieved with a primary fuel proportion of ~30%. This is due to the full use of air : with a low primary fuel proportion , its complete combustion occurs. With an increase in the primary fuel proportion above 30-40%, carbon gasification with carbon dioxide and water vapor begins already in the first stage, thus, the peak temperature decreases. On the other hand, the primary fuel underburning increases, which leads to an increase in the effective excess of oxidant (this is associated with an increase in the temperature of the product gas and a decrease in cold gas efficiency with a high primary fuel proportion).

The effect of the transition from the two-stage to the single-stage reactor operation can be observed in Fig. 6 (the curves were obtained for the conditions without secondary steam, however, the same picture is observed for other distributions of steam by stage). An abrupt change in temperature over a length of 3 m is associated with the transition of gas from one stage to another: in this case, the gas is cooled by heating and drying the secondary fuel. Then, heterogeneous chemical reactions begin: under small primary fuel proportions , the presence of residual oxygen leads to the development of exothermic reactions, and temperature profile can have a drop.

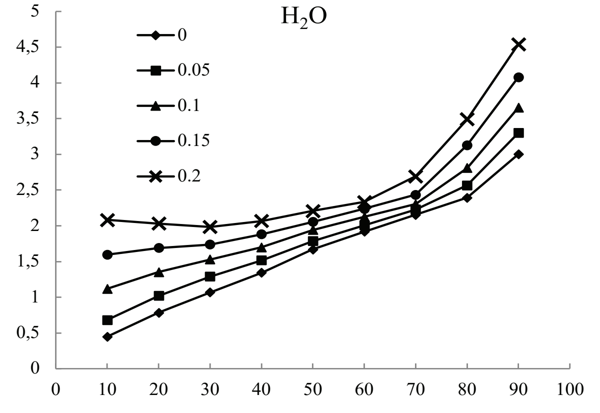

Fig. 12. The H_2O volume fraction in produced gas versus primary fuel and secondary steam proportions.

With a primary fuel proportion above 20–30%, there is no molecular oxygen in the products of the first stage. Therefore, the temperature curves are monotone, becoming the profile of a single-stage process with a high primary fuel ratio. This is evidenced by the course of changes in the intermediate gas composition and the first-stage oxidizer proportion α_1, as shown in Figs. 7-8 (calculations are carried out without secondary steam). With a high primary fuel proportion , the first stage produces the main quantity of combustible gases. Thus, with a primary fuel proportion of less than 30–40%, the operating conditions are characterized by an obvious two-stage temperature profile, and with a higher primary fuel proportion , the process gradually approaches the usual single-stage gasification process.

It is worth noting that the fraction of raw coal in the mixture with coke-ash residue entering the first stage (Fig. 5) remains almost constant when the proportion of the primary and secondary steam changes. Apparently, this characteristic of the process is determined primarily by the overall stoichiometry for the system of two reactors.

different behavior is observed before and after the primary fuel proportion of 60-70%. As already mentioned, above these values, a significant amount of underburning begins to form in the first stage, which leads to the present picture. This boundary can serve as the upper limit of the primary fuel proportion when choosing the operating conditions of a staged gasifier. The estimates obtained using one-dimensional model can be used as source data for CFD calculations [55]. According to the results of

3D-modeling, the efficiency of the gasification process with a primary fuel proportion of 30% is 84.9%. This value is in good agreement with the results of this model (84.5%). Further areas of research will be associated with the search for new ways to control the staged gasification processes. To this end, it is necessary to investigate the effect of heat and material flows between the stages and within them. The calculations show that an increase in the fraction of recycled coke-ash residue is a negative factor in general, therefore, this value should be minimized.

However, There can be various options of the process organization , that could shift boundaries of the efficient conditions (compared with those obtained in present work). For example, implementation of the additional stages: additional supply of gasification agent and fuel (including separation of the raw coal and coke-ash residue inputs), as well as the use of hot recycling systems for the coke-ash residue.

V. Conclusion

Alteration in the proportion of primary and secondary fuels in a two-stage pulverized gasification process leads to a successive change from the conditions with a staged sharp temperature profile (with a primary fuel proportion of 10-40%) to the conditions with a smoother profile that are typical of one-stage processes.

The calculated cold gas efficiency of the gasification process is quite close to the equilibrium (85.7%) in the range of the primary fuel proportion of 10-50%, the higher this proportion the lower the efficiency due to the formation of a significant amount of unburnt coke-ash residue. The use of secondary steam is ineffective in all the conditions considered. An analysis of the results makes it possible to choose the conditions in which the first stage implements the most complete fuel combustion: these conditions are primary candidates to be tested as optimal ones.

Further study can be focused on a new staged gasification process, including additional reagent supply sections (for example, an additional section for steam blast, separation of raw fuel and coke-ash residue, etc.)

Acknowledgements

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