A Numerical Study of the Influence of Process Parameters on the Efficiency of Staged Coal Gasification Using Mixtures of Oxygen and Carbon Dioxide

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Abstract — The paper considers a staged conversion process of pulverized coal fuel in the MHPS-type gasifier, which uses mixtures of oxygen and carbon dioxide as a gasifying agent instead of air. Similar conversion processes can be applied in the process diagrams with the capture and disposal of carbon dioxide. The research tool is a reduced-order mathematical model of coal particles’ conversion in a reacting gas flow. Replacement of nitrogen with carbon dioxide leads to significant changes in the gasification process characteristics: the average reaction temperature decreases, but this decrease is partially compensated by an increase in the concentration of gaseous reactants. Thus, the gasification process efficiency and the fuel conversion degree increase. Calculations make it possible to identify a range of parameters with the highest cold gas efficiency values. The influence of oxygen concentration is estimated, the dependence of the fuel conversion degree on the reaction temperature is analyzed.

Index Terms: coal gasification, staged process, mathematical modeling, oxy-fuel.

I. INTRODUCTION
Coal is the first candidate for displacement in the process of decarbonization of the energy sector. Despite its prevalence, large reserves, and low cost compared to other fossil fuels, coal has the worst forecast indicators [1, 2]. This fact, among other things, is due to the growing environmental requirements for energy technologies. Clean coal technologies, including gasification ones, are not yet widespread for various reasons, although their potential cannot be considered exhausted [3].

Gasification of coal dust and coal-water suspensions is widely used in industry, for example, in the production of chemicals [4]. According to [5], most of the large gasifiers operate for the needs of the chemical industry (mainly in China). There are some demonstration and commercial combined cycle power plants with integrated coal gasification (IGCC) [6]. Despite the better environmental characteristics and higher efficiency of such power plants, they are not widely used in the energy sector. This is due to both the progress of traditional coal-fired power plants operating with high steam parameters [7] and some unsolved problems (such as high capital costs [8] and insufficient reliability in comparison with other plants [9]). The IGCC efficiency largely depends on the efficiency and reliability of the gasification reactor and associated equipment (high-temperature heat exchangers, gas cleaning systems, and others). When the process conditions change, for example, fuel composition or power load, the reactor must maintain operation stability under existing technological constraints. The gasification agent composition and flow rate can be used as control parameters to this end.

Most of the existing high-power gasifiers use oxygen with a purity of up to 95% as a gasification agent [5]. In this case, the air separation costs should be compensated by an increase in the fuel conversion and the heating value of the produced gas. Other gasifier types use gasification agents with a low oxygen concentration (at the air level) [10]. In this case, the fuel conversion degree could be low. Consequently, additional systems are required for its combustion.

The flow diagrams of thermal power plants with carbon dioxide capture and storage (CCS) include the units designed to purify carbon dioxide from combustion products or intermediate products. The resulting carbon dioxide can be used, inter alia, to control the characteristics of combustion and gasification processes. For example, the recirculation of carbon dioxide and water vapor mixtures can be used in coal combustion to simplify the operation...
of CCS systems [11]. Similar processes were proposed for coal gasification in [12, 13].

In [14], the authors used a mathematical model of a stationary gasification process to compare the efficiency of coal-water and coal-carbon dioxide suspensions. Calculations show that carbon dioxide additives make it possible to reduce the specific oxygen consumption at a given temperature (in comparison with the coal-water suspension). However, the average fuel conversion degree decreases. The authors of [15] experimentally investigated the properties of carbon dioxide suspensions: measurements indicate that the viscosity and surface tension of carbon dioxide suspensions are lower than those of coal-water suspensions, which is why their transport and spraying require lower mechanical costs. They obtained carbon dioxide suspensions with a solid phase fraction of up to 66% of the mass. The work with carbon dioxide suspensions requires higher pressures. Therefore, their economic efficiency is determined by the operating pressure and thermal power of the reactor.

The process of staged gasification with carbon dioxide added (up to 25 vol. %) was proposed in [12]. The experimental setup made it possible to achieve cold gas efficiency (CGE) of about 45-50% with a fuel conversion degree of 90-95%. The low efficiency of the process is associated with high heat losses (the reactor capacity was three t/d, and the oxygen concentration was close to air). A similar process was considered in the study [12], which indicates that the recirculation of carbon dioxide and its partial use in the gasification process can increase the power plant efficiency. The studies [13, 16] proposed the flow diagrams of power plants, in which carbon dioxide is used not only in the gasification process but also as a working medium in power generation: the authors believe that such power plants can have a gross efficiency up to 50%.

The fuel conversion patterns change slightly when using $O_2/CO_2$ mixtures in contrast to $O_2/N_2$ mixtures. Oxygen diffusivity changes significantly over the particle surface: it decreases in the carbon dioxide-rich region and increases in the carbon monoxide-rich region, which is formed during gasification [17]. The contribution of the fuel reaction with oxygen declines. Therefore, the average combustion rate of particles decreases: the oxidation completeness depends to a greater extent on burning combustible gases formed on the fuel surface in the gas volume [18, 19]. Due to the lower reactivity of carbon to $CO_2$ and a decrease in the reaction temperature, the influence of the kinetic features of heterogeneous reactions and fuel porous structure increases [20]. The pyrolysis stage could be intensified due to an increase in the heat capacity of a high-temperature gas mixture [21]; however, with a decrease in the oxygen diffusivity, the ignition and combustion stages slow down [22]. The study [23] indicates that an increase in the heat capacity of a gas mixture due to carbon dioxide added could lead to flame extinction. In [24-26], the investigation indicates the effect of recirculation of combustion products on the structure of a pulverized coal flame: when carbon dioxide and water vapor are added, the flame length and average temperature decrease, which leads to a significant redistribution of heat fluxes both in the combustion region and on the walls of the burner.

Thus, the effect of water vapor and carbon dioxide addition could influence the macrokinetics of solid fuel conversion in different directions. Thermodynamic calculations show that during coal gasification, there can be regions of parameters with high CGE corresponding to very low temperatures of gasification products (which can be achieved only with a huge reaction zone) [27]. Therefore, when optimizing the parameters of the gasification reactor, it is necessary to raise the process temperature by increasing oxygen consumption [28]. The achievable efficiency of gasification processes using $O_2/CO_2$ mixtures will be determined by the competition between temperature and concentration factors.

There are some papers on modeling two-stage gasifiers, which address recirculation and consider the possibility of simultaneously varying both stoichiometric ratio and fuel distribution by stage [29, 30]. In this work, the model developed in [30, 31] is applied to the study of coal gasification under oxy-fuel conditions (i.e., gasification in mixtures of $O_2/CO_2$). The use of reduced-order models makes it possible to carry out calculations for a wider range of conditions (compared with the previous works) and get enhanced estimations for optimal parameters.

II. MATHEMATICAL MODEL

The two-stage gasifiers use chemical quenching. In the first stage, the fuel is burned to achieve the most complete conversion and high temperatures. The second stage suggests the supply of secondary fuel reacting with the decomposition products of primary fuel, which results in a decrease in the process temperature. The resulting coke ash residue is pneumatically transported back to the first stage for combustion. This diagram is currently used at Nakoso IGCC with Mitsubishi Hitachi Power Systems (MHPS) gasifier.

A mathematical model described in detail in [31, 32] is used to assess the characteristics of the gasification process. A feature of the model is the combination of computational approaches to chemical thermodynamics and kinetics of heterogeneous reactions, which can significantly reduce the computation efforts. Kinetics of heterogeneous reactions (drying, pyrolysis, gasification) is described using a system of differential equations, and the system composition is determined by the equilibrium conditions in the gas phase, i.e., by solving the extremum search problem [33]. Similar models were used to simulate coal gasification processes in [34-40].

The heat balance for fuel particles is written in the form of a differential equation:
\[ U_c \left( m \frac{dp}{dz} \right) = \alpha S_p \left( T_p - T_{rdz} \right) + \varepsilon \sigma S_p \left( T_p^4 - T_{rdz}^4 \right) + \sum_{j=1}^n Q_{j, p} \]  

(1)

In equation (1) \( z \) is the spatial coordinate (length of the reaction zone), \( m \); \( U \) is the particle velocity, m/s; \( m_p \) is the particle mass, kg; \( T_p \) is the particle temperature, K; \( \varepsilon \) is the specific heat capacity of fuel, J/(kg·K); \( \alpha \) is the heat transfer coefficient, W/(m²·K); \( S_p \) is the outer surface of the particle, m²; \( \varepsilon \) is the emissivity of the particle surface; \( \sigma \) is Stefan-Boltzmann constant, W/(m²·K⁴); \( T_b \) is the gas temperature, K; \( T_w \) is the wall temperature, K; \( r_j \) is the rate of the physicochemical process associated with the particle, kg/s (drying, pyrolysis, reactions with gases); \( Q_j \) is the thermal effect, J/kg (it is estimated from thermochemical data).

The drying rate \( r_{dr} \) is calculated using two formulas depending on the temperature conditions:

\[ r_{dr} = \begin{cases} \frac{\beta_{sl} M_w \chi_{H_2O} \left( P_{eq} - P_{H_2O} \right)}{R_g T_p}, & T_p \leq T_b, \\ \alpha S_p \left( T_p - T_{wall} \right) + \varepsilon \sigma S_p \left( T_p^4 - T_{wall}^4 \right), & T_p > T_b. \end{cases} \]  

(2)

Here \( T_b \) is the boiling point of water under the given conditions, K; \( \beta \) is the mass transfer coefficient, m/s; \( M_w \) is water molar mass; and \( \chi_{H_2O} \) is the partial pressure of water vapor, Pa; \( R_g \) is the universal gas constant, J/(mol·K); \( Q_{ass} \) is the water evaporation heat, J/kg.

The pyrolysis rate \( r_{pyr} \) depends on temperature according to the Arrhenius law:

\[ r_{pyr} = k_{pyr} \exp \left( -\frac{E_{pyr}}{R_g T_p} \right) m_p. \]  

(3)

Here \( k_{pyr} \) is the preexponential factor, 1/s; \( E_{pyr} \) is the activation energy, J/mol; \( m_p \) is the mass of volatile substances in the particle, kg. Heterogeneous reactions rate follows diffusion kinetics equation:

\[ r = \frac{S_p C_g}{k_{pyr}} \left( \frac{d_p}{\varepsilon_{try}} + N_{H_2} D_g \right). \]  

(4)

Here \( C_g \) is the concentration of gaseous oxidizer (O₂, CO₂, H₂O), \( k_{pyr} \) is the preexponential factor of heterogeneous reaction, m/s; \( E_{pyr} \) is the activation energy, J/mol; \( N_{H_2} \) is the diffusion Nusselt number; \( D_g \) is the diffusivity of gaseous oxidizer, m²/s; \( d_p \) is particle size, m. The solution to the system of equations for individual stages is found as follows. Differential equations (2)–(4) related to the kinetics of heterogeneous chemical transformations are solved at a given temperature distribution, while the gas composition is determined from the partial thermodynamic equilibrium problem [33]. Then the stationary problem of heat transfer is solved given heat sources (1). These steps are repeated until the resulting solution stops changing with iterations.

The numerical method of solving the problem for both sequential stages also involves iterations; the shutdown condition is the achievement of the specified accuracy in terms of the consumption of the recirculated coke-ash residue (5% of the total mass flow of the fuel entering the first stage). A two-stage gasifier is a system with feedback. Therefore, the existence of a stationary solution, in this case, is not obvious: even simple systems can exhibit complex dynamic behavior [41]. Preliminary calculations indicate that consumption of the recycled residue converges to a constant value, and this convergence is achieved rather fast: the determination of the stationary state parameters requires fewer than ten iterations (in most cases, fewer than five iterations). Nevertheless, the question of the number and stability of stationary states, generally speaking, remains open and should be solved employing appropriate models [42].

Our previous research [43] investigated the effect of adding carbon dioxide and water vapor in the oxygen-blown one-stage coal gasification process. This work focuses on a staged process with an oxygen concentration of 21-30%. The initial gasification agent temperature is 500 K. The geometric parameters of reactors are constant for all calculations: the reaction zone length is 20 m (including the length of the first stage equal to 3 m), the diameter is 3 m. Fuel consumption is 1700 t/d, average particle size is 100 μm. Fuel composition is as follows: moisture content is 9.2 wt%; ash content is 12.7 wt%; volatiles yield is 30.9 wt%; carbon content is 82.3 wt%; hydrogen content is 5.06 wt%; oxygen content is 9.47 wt%; nitrogen content is 1.91 wt%; sulfur content is 1.09 wt%. The blast is distributed between the stages as follows: 90% goes to the first stage, and 10% goes to the second stage. Solid particles do not pass from the first stage to the second: it is assumed that the fuel is either completely burned or escapes the reactor with slag as mechanical underburning. Recirculation of excess fuel mass leads to computational difficulties (the iterative process may be convergent due to this approximation). We assume that after the second stage the coke-ash residue is cooled to input temperature before returning to the first stage.

### III. Modeling Results and Discussion

Calculated stationary profiles of temperature and gas composition during gasification of pulverized coal in \( O_2/N_2 \) and \( O_2/CO_2 \) mixtures are shown in Fig. 1 and 2 (the initial volume fraction of oxygen in both cases is equal to 21%). The reaction temperature decreases significantly in the case of the \( O_2/CO_2 \) mixture (by 300-400 K in the first stage and by 100-200 K in the second stage). However, due to the high concentration of \( CO_2 \), a more complete fuel conversion is observed, and the produced gas has a higher calorific value (similar results were obtained in [44] for one-stage processes). This implies that temperature declines not only due to an increase in the specific heat capacity of gas flow but also due to an intensification of endothermic reactions.
The CGE dependence on the fraction of primary fuel, when using the O$_2$/N$_2$ mixture with an oxygen concentration of 21%, is shown in Fig. 3. The maximum CGE corresponds to a primary fuel fraction of about 60%. Earlier [30], the value of the optimal primary fuel fraction obtained for gasification in high-temperature air (initial temperature 1173 K) was about 30%. In present calculations, the maximum gas temperature at the first stage outlet is reached at a primary fuel fraction of 30-40%.

With a primary fuel fraction of 60% and a stoichiometric ratio of 0.4, the gas temperature is about 1900 K, which is permissible under slagging conditions, and allows the process in the second stage with a sufficiently complete conversion of the secondary fuel. A further increase in the primary fuel fraction leads to a decrease in the temperature, and fuel conversion becomes low.

Switching from air blast to mixtures of oxygen and carbon dioxide, on the one hand, increases the concentration of oxidants, on the other hand, decreases the average reaction temperature. Therefore, the efficiency of fuel gasification under oxy-fuel conditions is determined by the ratio of these opposing factors.

The primary fuel dependence of CGE for O$_2$/CO$_2$ mixtures with oxygen concentration equal to 21% is shown in Fig. 4. The optimal fraction of primary fuels is still about 60%. The form of the dependence does not change, optimal stoichiometric ratio value, however, shifts towards lower values. At a stoichiometric ratio equal to 0.45, a monotonic decrease in CGE is observed with an increase in the primary fuel fraction: such dependence [30] was observed for the high-temperature air gasification process. Under small proportions of primary fuel (up to 30%), the optimal value of the stoichiometric coefficient is 0.4, as for air gasification. With a further increase in the primary fuel fraction, the optimal value of the stoichiometric ratio goes down to 0.35. It can be assumed that this is due to a change in the dominant stage. With small proportions
Fig. 4. Cold gas efficiency of two-stage gasification process with \( \text{O}_2/\text{CO}_2 \) mixture (oxygen volume concentration is 21%): dependence on stoichiometric ratio and primary fuel fraction.

Fig. 5. Cold gas efficiency of two-stage gasification process with the \( \text{O}_2/\text{CO}_2 \) mixture (oxygen volume concentration is 25%): dependence on stoichiometric ratio and primary fuel fraction.

Fig. 6. Cold gas efficiency of two-stage gasification process with the \( \text{O}_2/\text{CO}_2 \) mixture (oxygen volume concentration is 30%): dependence on stoichiometric ratio and primary fuel fraction.

Fig. 7. Dependence of the maximum cold gas efficiency on the gasification agent composition and the stoichiometric ratio (numbers in legend).

of the primary fuel, gasification of the secondary fuel is of decisive importance; therefore, to increase the process efficiency, it is necessary to increase the temperature of hot gases before the second stage. With large proportions of secondary fuel, the completeness of the primary fuel conversion into combustible gas becomes more important; the secondary fuel is needed only for chemical quenching. Similar competition between the process stages was observed in the previous study [30] that examined the transition between one- and two-stage gasification conditions.

The same qualitative regularities are observed for the gasification process in mixtures with an oxygen volume concentration of 25 and 30% (Figs. 5 and 6). The CGE dependence on the primary fuel fraction flattens already at a stoichiometric ratio equal to 0.4. An oxygen concentration increase leads to a rise in the reaction temperature, therefore, at an oxygen volume concentration of 30%, the optimal value of the stoichiometric coefficient is 0.35 for all fuel distribution alternatives by stage. The temperature rise facilitates the endothermic reaction of fuel carbon with carbon dioxide; therefore, the average conversion rate and maximum chemical efficiency increase with the growing oxygen concentration.

The values of the maximum CGE are shown in Fig. 7. The higher the oxygen concentration, the higher the gasification process efficiency. The optimal stoichiometric ratio varies in a range of 0.3-0.4. With the air gasification of coal, one can reach a CGE of about 75%. With the \( \text{O}_2/\text{CO}_2 \) mixtures, even at equal oxygen concentrations, the CGE can be increased up to 78%. The maximum CGE in all the considered options is about 82% (at an oxygen volume concentration of 30%, a stoichiometric ratio of 0.35, and a primary fuel fraction of 50%).

The temperature dependences of the fuel conversion degree for the first and second stages (based on the results of all calculations) are shown in Fig. 8. One can see that
for \(O_2/CO_2\) mixtures, the points are grouped around a common curve. Small scattering arises due to different conditions, mainly stoichiometric and kinetic factors (for example, the instability of combustion in the first reactor or a deficiency of primary fuel). Air gasification requires higher temperatures to achieve a complete fuel conversion: apparently, the concentration factor is prevailing.

IV. conclusion

The calculations show that in the process of pulverized coal fuel staged gasification, the efficiency can be increased with the mixtures of oxygen and carbon dioxide used as a gasifying agent. The magnitude of the effect is determined by oxygen concentration and process temperature constraints. The optimal fraction of primary fuel is in a range of 50–60% for all cases, and the optimal value of the stoichiometric ratio is 0.35–0.4. The gasification process CGE rises from 75 to 82% with an increase in the oxygen volume concentration from 21 to 30%.

The results obtained can be used to assess the efficiency of gasification reactors and power plants with carbon dioxide capture and storage systems.

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