Mathematical modelling of a staged pulverized coal gasification using \( \text{O}_2/\text{CO}_2 \) mixtures

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Abstract. Conversion of pulverized coal in a two-stage gasifier is studied. When considering carbon capture power plants, mixtures of oxygen with carbon dioxide may be used as a gasification agent. Carbon dioxide is a gasification agent, so characteristics of the gasification process change significantly compared to gasification in oxygen-nitrogen mixtures. The conversion efficiency is determined by the thermophysical factor (change in the heat capacity of the gas mixture and transfer coefficients) and the concentration factor (increase in the concentration of the gaseous reagent). The ratio of primary and secondary fuel consumption determines the leading stage of the process. The influence of process efficiency on oxygen concentration is estimated in the range of 21–30 vol. %.

1. Introduction
Gasification of coal dust and coal-water suspensions is widely used in chemical industry for producing hydrogen and syngas [1]. There are several demonstration and commercial combined cycle power plants with integrated coal gasification (IGCC) [2], however, despite better environmental characteristics and higher efficiency, such power plants are not yet widely used in the energy sector due to such issues as high capital cost and lower reliability compared with traditional coal-fired plants [3]. Gasifier is a core unit of IGCC plant, so its operation determines, to a significant extent, efficiency and reliability of the whole plant.

Industrial coal gasifiers usually consume oxygen with a purity up to 95% in order to produce gas with high fraction of \( \text{CO} \) and \( \text{H}_2 \). In the case of energy use of produced gas, one can use gasification agent with an oxygen concentration at the air level [4]. To ensure the complete conversion of carbon, air gasifiers include coke-ash residue recirculation systems. A well-known gasifier of this type is the MHPS-gasifier (Mitsubishi and Hitachi Power Systems) [5].

Promising schemes of coal-fired plants with capture and storage of carbon dioxide (CCS) include units for purification of carbon dioxide from combustion products or intermediate products [6]. The purified carbon dioxide may be used at the plant, for example, to control the characteristics of combustion and gasification processes. The process of staged gasification with the air-carbon dioxide mixtures was proposed in [7]. The experimental setup allowed achieving cold gas efficiency (CGE) of about 45-50% with a fuel conversion degree of 90-95%. A similar process was considered in [8], where it was shown that recirculation of carbon dioxide and its partial use in the gasification process can increase the power plant efficiency. Carbon dioxide may be used not only at gasification stage [9, 10], but also as a heat carrier and working medium in the power cycle [11].

In this work, the model of pulverized fuel gasification [12, 13] is applied to the study coal conversion in mixtures of \( \text{O}_2/\text{CO}_2 \). A feature of the model is the combination of computational
approaches of chemical thermodynamics and heterogeneous reactions kinetics, which allows significantly reducing the computational cost, so we can vary stoichiometric ratio and fuel distribution in a wide range [12].

2. Brief model description

The two-stage gasifiers use chemical quenching. In the first stage, the fuel is burned to achieve the most complete conversion and high temperatures. In the second stage, secondary fuel is supplied; it reacts with the decomposition products of the primary fuel, as a result of which the process temperature decreases. The resulting coke ash residue is pneumatically transported back to the first stage for combustion. To assess the characteristics of the gasification process, a mathematical model is used; it is described in detail in [12, 14]. The kinetics of heterogeneous reactions (drying, pyrolysis, gasification) is described using a system of differential equations, and the composition of the system is determined by the equilibrium conditions in the gas phase, i.e., as a result of solving the extremum search problem [15]. Similar models were used to simulate coal gasification processes in papers [16–22].

The numerical solution of the problem for both sequential stages is also carried out iteratively. 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. However, preliminary calculations have shown that the flow rate of the coke-ash residue converges to a constant value, and this convergence is achieved rather quickly: to determine the parameters of the stationary state, less than 10 iterations are required (in most cases, less than 5 iterations).

The initial gasification agent temperature is 500 K. The geometric parameters of the reactors are constant for all calculations: the length of the reaction zone is 20 m (of which the length of the first stage is 3 m), and the diameter is 3 m. Fuel consumption is 70.8 t/h, and average particle size is 100 μm. Fuel composition is as follows: moisture content is 9.2 wt.%; ash content is wt. 12.7%; 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.%; and sulfur content is 1.09 wt.%. The gasification agent 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. After the second stage the coke-ash residue is cooled to input temperature before returning to the first stage.

3. Modelling results

The main efficiency criterion of our interest is cold gas efficiency (CGE):

$$\eta = \frac{G_f (y_{CO}Q_{CO} + y_{H_2}Q_{H_2} + y_{CH_4}Q_{CH_4})}{G_f Q_f} \times 100\% .$$

Here $G_f$ is the gas output flow rate, $G_f$ is the input coal flow rate, $Q_f$ is the coal heat value, and $y$ is the mass fraction of combustible components in produced gas.

The dependence of the CGE when using O$_2$/N$_2$ mixture with an oxygen concentration of 21 vol.% on the fraction of primary fuel is shown in Fig. 1a. The maximum CGE corresponds to a primary fuel fraction of about 60 wt.%. Earlier in paper [12] we obtained the value of the optimal fraction of primary fuel of about 30 wt.% for gasification in high-temperature air (initial temperature of 1173 K). In present calculations the maximum gas temperature at the outlet of the first stage is reached at primary fuel fraction of 30–40 wt.%. With a primary fuel fraction of 60 wt.% and a stoichiometric ratio of 0.4, the gas temperature is about 1900 K, which is permissible in relation to the conditions of slagging and allows the process to be carried out at the second stage with a rather complete conversion of the secondary fuel. A further increase in the fraction of primary fuel leads to a decrease in temperature, and fuel conversion becomes low.
When changing gasification agent from air to O$_2$/CO$_2$ mixtures, on the one hand, an increase in the concentration of oxidants occurs, and on the other hand, average reaction temperature decreases. Therefore, the efficiency of fuel gasification under oxyfuel conditions is determined by the ratio of these opposing factors.

The dependence of the CGE for O$_2$/CO$_2$ mixtures with oxygen concentration equal to 21 vol.% is shown in Fig. 1b. The optimal fraction of primary fuels is still about 60%. The form of the dependence does not change; however, the value of the optimal stoichiometric ratio shifts towards lower values. At stoichiometric ratio equal to 0.45, a monotonic decrease in the CGE is observed with an increase in the primary fuel fraction. 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 decreases to 0.35. It may be assumed that this is due to a change in the dominant stage. With small proportions of the primary fuel, gasification of the secondary fuel is of decisive importance; therefore, in order to increase the efficiency of the process, it is necessary to increase the temperature of the 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; secondary fuel is needed only for chemical quenching. A similar competition between the stages of the process was observed in previous work [12], where we studied the transition between one- and two-stage gasification conditions.

**Figure 1.** Cold gas efficiency of staged gasification process (oxygen concentration is 21 vol. %): dependence on stoichiometric ratio and primary fuel fraction.

**Figure 2.** Dependence of the maximum CGE on the gasification agent composition and the stoichiometric ratio.
The dependence of CGE on stoichiometric ratio usually has an extremum [23]. The values of the maximum CGE are shown in Fig. 2. The optimal stoichiometric ratio for all cases varies in the range of 0.3–0.4. With the air gasification of coal, it is possible to achieve CGE of about 75%. With the use of O2/CO2 mixtures, even at equal oxygen concentration, the CGE may be increased up to 78%. The maximum CGE among all the considered options is about 82% (at oxygen volume concentration of 30 vol.%, stoichiometric ratio of 0.35, and primary fuel fraction of 50 wt.%).

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

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